Activity and Chemical Composition Modifications Induced by Spallation Residues in a Pb or Pb-Bi Target*

L. Donadille, A. Boudard, J.C. David, S. Leray and C. Volant

DAPNIA/SPhN, CEA/ Saclay, 91191 Gif-sur-Yvette cedex, France
sleray@cea.fr

Abstract -*Calculations of the chemical composition modifications and activity induced by spallation residues in thick Pb or Pb-Bi targets, after 1 year of irradiation by 1 GeV protons, have been performed with LAHET3, in which the new INCL4-ABLA model has been implemented. Since this model has been shown to fairly reproduce recent measurements of the isotopic distributions of spallation residues produced in Pb by 1 GeV protons (performed with the reverse kinematics technique) and evolution with energy of the production of radioactive isotopes (measured by γ-spectrometry), it is now possible to rely on the prediction of the calculation and assess the uncertainties on the predictions. It is found that for activity estimations, the use of the new models instead of standard ones (as the Bertini-Dresner combination) changes only little the prediction. However, for chemical impurities up to a factor 3 discrepancies can be found with the new model, in particular in the case of some volatile elements, due to a bad prediction of the fission fragment yields in the standard codes.*

I. INTRODUCTION

In an Accelerator-Driven System (or a spallation neutron source) spallation target, a large variety of isotopes is produced by spallation reactions in addition to those created by activation by the low energy neutron flux. A lot of them are radioactive and could be a source of concern for radioprotection. For instance, it is important to know the time evolution of the target activity to determine when it is possible to approach for maintenance or unplanned intervention. Long-lived isotopes are produced which will be responsible for the long-term radiotoxicity of the target after operation. Also, the chemical modifications due to the growth of impurities could lead to corrosion problems on the structure materials in contact with the liquid metal.

Up to recently, little confidence could be granted to the predictions of spallation residue production by high-energy models [1]. Complete isotopic production cross-sections of residues in Pb at 1 GeV [2, 3] are now available, thanks to the GSI (Darmstadt) experiments using the reverse kinematics techniques. These data showed that the high-energy models used in standard high-energy transport code, in fact the Bertini-Dresner [4] combination, did not correctly predict in particular the isotopic distributions and the fission fragment production [5]. On the other hand, an important effort has been devoted to the building of new improved physics models that could more reliably predict all quantities necessary for the design of spallation targets. The INCL4 intranuclear cascade coupled to the ABLA evaporation-fission model [6, 7] (see also A. Boudard et al. in this conference) is an example of such new codes that have been shown to give a satisfying agreement with many experimental observables, in particular concerning the isotopic distributions of spallation residues in the region of fission and heavy evaporation products. These models have been recently implemented into the LAHET3 code system [8] and in MCNPX [9]. It is therefore now possible to calculate with these codes quantities like the activity due to the spallation residues or chemical impurity production for real spallation targets with an improved confidence. In this paper we present such calculations and show comparisons with results of standard codes, as those obtained for instance by Shubin et al. [10].

II. CALCULATION METHOD

Calculations have been made for a 10 cm radius, 1 m long target, supposed to have been irradiated by a 1 GeV, 1 mA proton beam during one year. Both Pb and Pb-Bi targets have been investigated, but we show mostly Pb-Bi calculations in this paper since part of the Pb results have already been published in [11]. Spallation residue production due to the primary interactions and secondary neutron (down to 20 MeV) and proton (down to stopping) induced reactions has been estimated using the LAHET3 high-energy transport code. Two different options for the models that generate the elementary cross-sections were studied: either the (standard) Bertini-Dresner or the new INCL4-ABLA combination of intranuclear cascade-deexcitation models. The evolution of the nuclides concentrations as a function of time has been calculated with the ORIHET3 [12] decay code. The neutrons below 20 MeV were transported by MCNP4C. To evaluate the activation due to the resulting neutron flux, the DARWIN [13] code (with 33 energy groups) was used. We assumed a constant target composition, as it was checked that the activation of the spallation products should be negligible. Since in an ADS or a spallation source the target will also be subject to an external neutron flux from the sub-critical core or the moderator/reflector, a calculation has also been performed with a target surrounded by D_2O, in order to estimate this contribution.

III. CALCULATIONS OF ACTIVITY

III.A.The Different Contributions to the Activity

Fig.1 shows the results of the calculations of the different contributions to the activity in the case of a Pb-Bi target, as a function of the cooling time. The main contribution comes from the spallation products (solid line) and is almost two orders of magnitude larger than the one due to activation by the low energy neutron flux (dotted line) up to one hundred years. Even in the case of a target surrounded by D_2O (dashed-dotted line), leading to a total neutron flux of 3×10^{14} $n.cm^{-2}.s^{-1}$, the spallation products always dominate the activity except between one month and one year when the activation of ^{209}Bi, leading to ^{210}Bi and then ^{210}Po which have respective half lifes of 5 and 138 days, is of the same order of magnitude. This calculation was done using the Bertini-Dresner model.

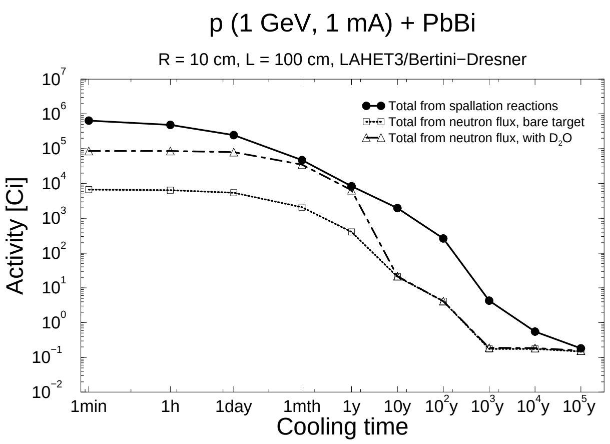

Fig. 1. Activity (in Curies) of a Pb-Bi target as a function of time, after one year of irradiation, due to the spallation products (solid line), activation by the neutron flux below 20 MeV in the case of a bare target (dotted line) or in the case of a target surrounded by D_2O (dashed-dotted line).

As far as the spallation products are concerned, a large number of isotopes are actually contributing to the total activity. During the irradiation phase (not shown), the total activity almost saturates at 5×10^5 Ci after about one month. At this time and up to one day after irradiation, the main contributors to the activity are ^{206}Bi and ^{205}Bi for both the Bertini-Dresner and INCL4-ABLA calculations. For the decay stage, shown in fig.2 in the case of the INCL4-ABLA calculations, it can be seen that the contribution of these two nuclides is less than 10% of the total activity. This means that a large number of different isotopes contribute almost equivalently to the total activity. A similar behavior is observed up to almost a month of decay, in fact because most of these nuclides have rather short periods ($T_{1/2}$ < 30 d). For longer decay times the activity is due to few long-lived nuclides only. For example, after 10 years of decay the nuclide ^{207}Bi represents 60% of the activity, and after 10^4 years the dominant nuclide, representing 50% of the activity, is ^{202}Tl, populated by the beta-decay of the long-lived ^{202}Pb. It can be noticed that only heavy residues close to the target elements contribute significantly. The activity due to the fission products is always less than 10-15% of the total. These results are in good agreement with those of Shubin et al. [10].

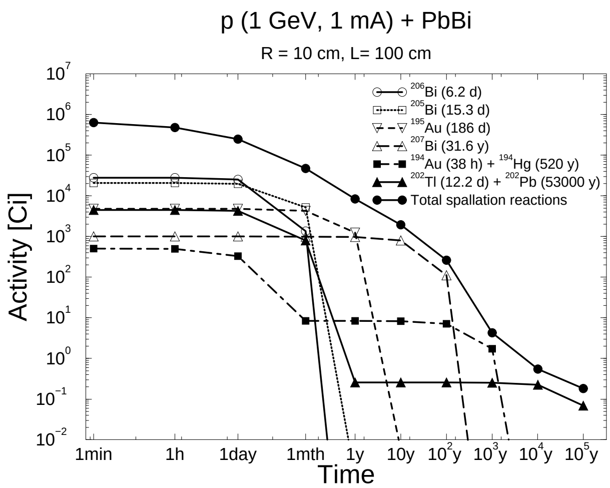

Fig. 2. Main contributors to the activity (in Curies) of a Pb-Bi target as a function of time, after one year of irradiation, calculated with the LAHET3 code system using the INCL4-ABLA models.

III.B. Comparison of Different Model Predictions

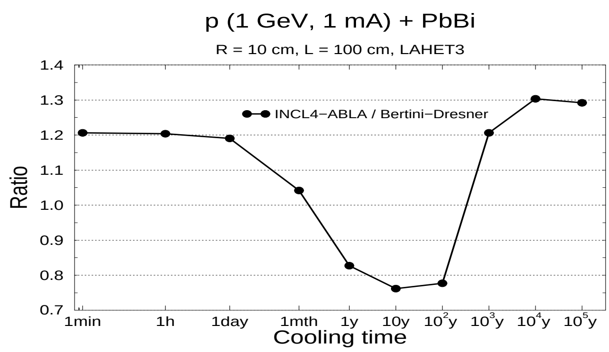

Fig. 3. Ratio of the activity due to the spallation products calculated using the INCL4-ABLA and the Bertini-Dresner models, for a Pb-Bi target, as a function of time after one year of irradiation.

When Bertini-Dresner is used instead of INCL4-ABLA, the results are globally the same although the relative contributions of individual isotopes may be different. In fig.3, we show the ratio between the activity calculated with INCL4-ABLA and the one with Bertini-Dresner as a function of the decay time. It can be seen that maximum differences are of the order of ±30%. This is not surprising since actually the models give similar results for residues very close to the target elements, with slight differences on the isotopic distributions, the larger discrepancies appearing for lighter isotopes. However, what is new is that we can now rely more confidently on these predictions because we know that the INCL4-ABLA does predict correctly the production of the involved isotopes [6]. Indeed, in a such thick target, the primary proton induces on average one high-energy and two secondary lower energy interactions. As already said, the model agrees totally with the isotopic production at 1 GeV and, as shown in fig.4 in the case of Bi isotopes produced in a natural lead target, reproduces quite well the energy dependence of the production cross-sections, measured by γ-spectrometry by Gloris et al. [14], down, at least, to a few tens of MeV. The discrepancies between the model and the data being at most 30-40%, a similar uncertainty can be assessd for the predicted activity. Similar conclusions could also be drawn for a thick lead target.

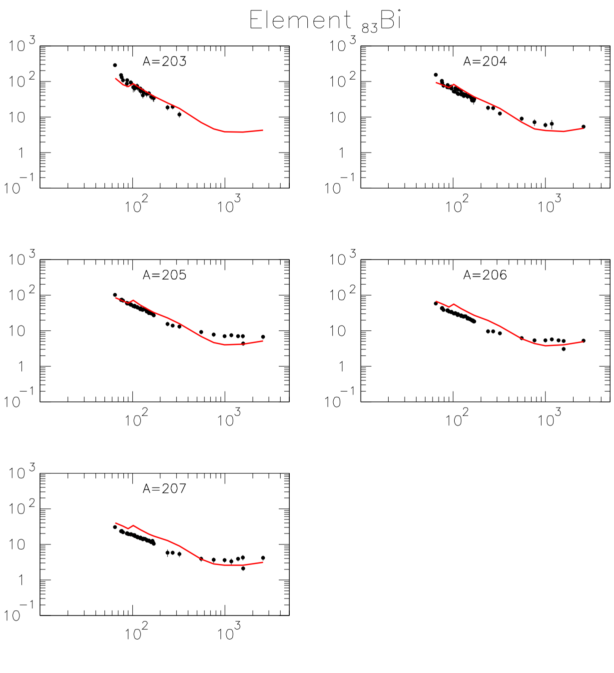

Fig. 4. Comparison of INCL4-ABLA predictions with Bi isotope production cross-sections (mb) from a natural Pb target as a function of incident proton energy (MeV) measured by Gloris et al. [14].

III.C. Comparison of Pb and Pb-Bi Targets

For the activity due to spallation residues, a similar behavior was found for the two targets: same level of total activity, contribution of a large number of isotopes even if there are not the same ones, main contributors being nuclei very close to the target elements. However, after around thirty years of decay, a factor 3 to 5 (depending on the models used) larger activity is predicted, due to a large production of ^{207}Bi, in the case of Pb-Bi. This can be seen in fig.5, which shows the ratio between the activity of the two targets calculated with either INCL4-ABLA or Bertini-Dresner as a function of the decay time. Together with the generation of ^{210}Po through activation by the neutron flux, irrespective of other technological considerations, this would favor the choice of Pb compared to Pb-Bi.

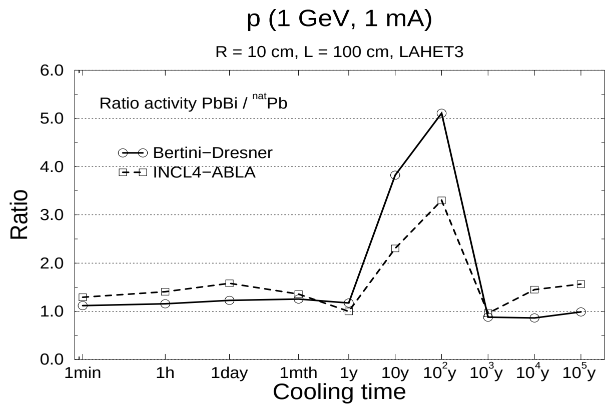

Fig. 5. Ratio of the activity due to the spallation products of a Pb-Bi and a Pb target, calculated using the INCL4-ABLA (dashed line) and the Bertini-Dresner (solid line) models, as a function of time after one year of irradiation.

IV. PRODUCTION OF IMPURITIES

As already stated, in a liquid metal target, chemical impurities produced by spallation reactions, can lead to corrosion problems on the container of the target or on the window if in contact with the metal. We have therefore calculated the concentrations of the different elements generated after one year of irradiation for a Pb and a Pb-Bi target. The results obtained with INCL4-ABLA (dotted curve) and Bertini-dresner (dashed-dotted curve) are shown in fig. 6. One can see fluctuations due to the fact that radioactive nuclei more often decay towards even atomic number. The ratio between the two calculations (solid line) is also shown. Here the discrepancy between the model can reach a factor 3 in the region of fission fragments and up to 30 for the very light evaporation residues.

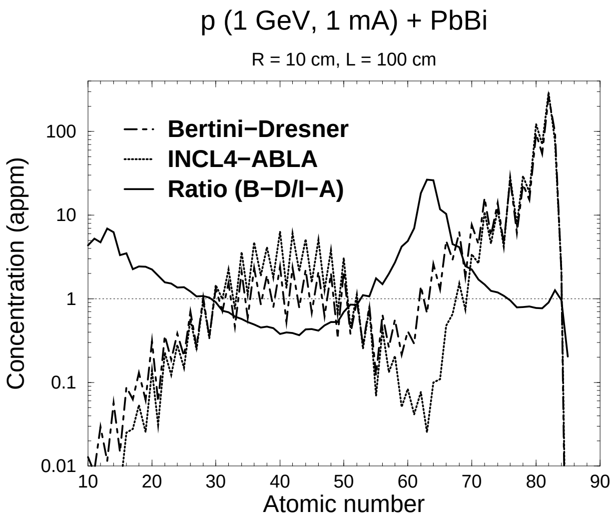

Fig. 6. Concentration in appm of the different chemical impurities produced by spallation in a Pb-Bi target after one year of irradiation, calculated with LAHET3 using the INCL4-ABLA (dotted curve) or the Bertini-Dresner (dashed-dotted curve) model and ratio between the two calculations (solid line).

Actually, this reflects exactly the differences in the elementary production of the fission fragments at 1 GeV as these fragments are produced only in relatively high-energy reactions. Since it has been established in [5, 6] that INCL4-ABLA predicts much better the fission region, as it can be seen in fig.7, this calculation is obviously an improvement compared to the standard codes. For the light evaporation residues that arise in very low concentrations, the situation is not as good since none of the models correctly predicts the elementary cross-sections but this gives an idea of the uncertainty on their production.

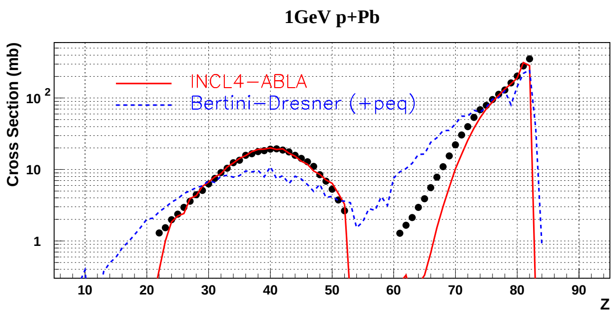

Fig. 7. Comparison of INCL4-ABLA predictions with the charge distribution of residues measured by the reverse kinematics technique at GSI [2, 3] for the reaction ^{208}Pb (1 GeV/A) + p.

Some of the fission products are volatile gases, of which some isotopes are radioactive and can be a concern for radioprotection in case of a containment failure. It is therefore interesting to investigate more precisely these elements. Table 1 gives the results from the two models, for the expected concentrations of krypton, iodine and xenon isotopes and the total for each element. For the latter, the ratio is the one found in fig.5 for Z=36, 53 and 54 respectively, and is a factor 2 maximum. However, for isotopic concentrations the discrepancies can reach a factor 4, due to the fact that the isotopic distributions are different in the two models. Here again, we can stress that with INCL4-ABLA we have a more reliable prediction but also that when using Bertini-Dresner the error on the prediction is certainly not larger than a factor 4 to 5.

Table 1. Comparison of INCL4-ABLA and Bertini-Dresner predictions for volatile gas production and ratio between the two calculations.

Isotope	half life	PbBi		
		Concentration (appm)		Ratio
		Bert-Dres	Incl4-Abla	I-A/B-D
81 Kr	2.3E+05 y	2,4E-01	4,0E-01	1,67
85 Kr	10,8 y	8,1E-02	2,8E-01	3,46
Kr		**2,3E+00**	**4,7E+00**	**2,04**
124 I	4,1 d	9,6E-04	1,2E-03	1,25
125 I	59,4 d	3,1E-02	3,4E-02	1,10
126 I	13,1 d	2,4E-03	2,9E-03	1,21
129 I	1.7E+07 y	6,2E-02	4,3E-02	0,69
131 I	8,0 d	9,1E-04	2,8E-04	0,31
I		**2,8E-01**	**2,5E-01**	**0,89**
127 Xe	36,4 d	1,5E-02	1,6E-02	1,07
129m Xe	8,9 d	3,8E-03	3,1E-03	0,82
131m Xe	11,9 d	5,6E-04	6,0E-04	1,071
133 Xe	5,2 d	6,9E-04	2,1E-04	0,30
133m Xe	2,2 d	1,9E-05	2,9E-05	1,526
135 Xe	9,1 d	2,0E-05	5,0E-06	0,25
Xe		**8,2E-01**	**7,7E-01**	**0,94**

IV. CONCLUSIONS

In this paper, we have studied the activity and chemical impurity production in realistic Pb and Pb-Bi spallation target. The emphasis has been put on spallation residues that represent the main contribution to the activity even in the presence of a reactor neutron flux. The results have been obtained with the new INCL4-ABLA spallation model implemented into LAHET3. Since this model has been validated on a large set of experimental data concerning both isotopic distributions at one energy and the energy dependence for the main contributors, we can claim that our predictions are more reliable than those done with standard high-energy transport codes are. However, sometimes the results of INCL4/ABLA and

Bertini/Dresner, in particular for the activity, are not so different. The main finding, in this case, is that we can now assess the uncertainty in the prediction with quite a good confidence. On the contrary, for chemical composition modifications and production of volatile gases, large differences have been found.

REFERENCES

1. R. MICHEL and P. NAGEL, *Int. Code and Model Intercomparison for Intermediate Energy Activation Yields*, OECD/NEA (1997).
2. W. WLAZLO et al., *Phys. Rev. Lett.* **84,** *5736* (2000).
3. T. ENQVIST et al., *Nucl. Phys.* **A686,** *481* (2001).
4. H.W. BERTINI, *Phys. Rev.* **131,** *1801* (1963); L. DRESNER, *ORNL Report ORNL-TM-196* (1962).
5. S. LERAY et al., *Proceedings of the Monte-Carlo 2000 International Conference*, *p1111*, A.Kling, F.Barao, M.Nakagawa, L.Tavora, Eds., Lisbon (2001).
6. A. BOUDARD et al., *Phys. Rev.* **C66,** *044615* (2002).
7. A.R.JUNGHANS et al., *Nucl. Phys.* **A629,** *635* (1998).
8. R.E. PRAEL and H. LICHTENSTEIN, *LANL Report LA-UR-89-3014* (1989) and R.E. PRAEL, private communication.
9. J.S. HENDRICKS et al., MCNPX 2.5.d, report LA-UR-03-5916.
10. Yu.N. SHUBIN, et al., Proc. Int. Conf. Evaluation of Emerging Nuclear Fuel Cycle System, GLOBAL 1995, Sept. 11-14 (1995).
11. L. DONADILLE et al., *J. of Nucl. Science and Techn.*, **Suppl. 2**, *p1194*, August 2002.
12. F. ATCHISON, H. SCHAAL, private communication.
13. T. TZILANITZARA et al., *J. of Nucl. Sc. And Tech.*, **Suppl. 1**, *845* (2000).
14. M. GLORIS et al., *Nucl. Instr. and Meth.* **A463,** *593* (2001).

* Work supported by the HINDAS (FIKW-CT-2000-00031) European Commission contract