IMPACT OF HIGH ENERGY NUCLEAR DATA ON THE DESIGN OF SPALLATION SOURCES*

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The HINDAS high-energy programme has provided a large amount of experimental data that has led to a better understanding of the spallation reaction mechanism and development of more reliable spallation models. These models, which have been implemented into high-energy transport codes, or sometimes directly the data, can be now used to predict with a larger confidence or, at least with a known uncertainty, some important quantities for the design of ADS spallation modules. In this paper, we present examples concerning the chemical impurities and activity induced by spallation residues in thick Pb-Bi target, after one year of irradiation by 1 GeV protons and chemical composition modifications and number of atom displacements (DPA) expected in an ADS window after one year of operation.

1 Introduction

Within the HINDAS high-energy programme a large amount of experimental data concerning neutron, charged particle and residue production have been collected. This has led to a better understanding of the spallation reaction mechanism and the development of more reliable spallation models, the INCL4 intranuclear cascade [1] and ABLA evaporation-fission [2] models, with known successes and remaining deficiencies. These models have recently been implemented into high-energy transport codes that can be now used to predict with a larger confidence or, at least with a known uncertainty, some important quantities for the design of ADS spallation modules. In this paper, we present examples concerning the impact of the measurements of isotopic cross-sections with the reverse kinematics method at GSI and excitation functions by mass- and γ-spectrometry on the predictions of activity, impurity production in a Pb-Bi target and composition modifications, DPAs in a steel window. Other studies related to neutron production and damage due to gas emission have also been conducted within HINDAS but are not presented here.

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2 Activity and chemical impurities in a Pb-Bi spallation target

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 are a source of concern for radioprotection. Long-lived isotopes are also produced which will be responsible for the long-term radiotoxicity of the target after operation. In addition, the chemical modifications due to the growth of impurities could lead to corrosion problems on the structure materials in contact with the liquid metal.

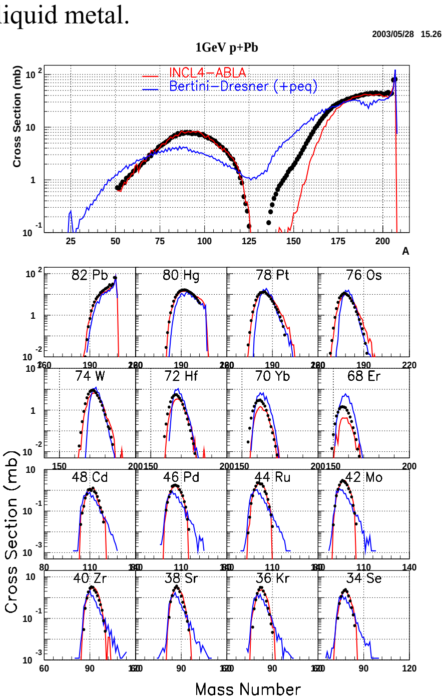

Figure 1. Mass and a few isotopic distributions of residues produced in Pb+p at 1 GeV, measured at GSI [2, 3], and compared to calculations with Bertini-Dresner or INCL4-ABLA models.

Up to recently, little confidence could be granted to the predictions of spallation residue production by high-energy models. Complete isotopic production cross-sections of residues in Pb at 1 GeV [3, 4] 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 [5] combination, did not correctly predict in particular the isotopic distributions and the fission fragment production [6]. The INCL4 intranuclear cascade coupled to the ABLA evaporation-fission model [1, 2] (see also Th. Aoust et al. in this conference) developed within HINDAS has 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 (see fig.1). These models have been recently implemented into the LAHET3 code system [7] and MCNPX [8]. 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 and compare with results of standard codes, as those obtained for instance by Shubin et al. [9].

2.1 *Calculation method*

Calculations have been made for a 10 cm radius, 1 m long Pb-Bi target, supposed to have been irradiated by a 1 GeV, 1 mA proton beam during one year. 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 [10] decay code. The neutrons below 20 MeV were transported by MCNP4C. To evaluate the activation due to the resulting neutron flux, the DARWIN [11] 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.

2.2 *Calculations of activity*

It has been shown in [12] that the main contribution comes from the spallation products and is almost two orders of magnitude larger than the one due to activation by the low energy neutron flux up to one hundred years. Even in the case of a target surrounded by D_2O, leading to a total neutron flux of $3x10^{14}$ $n.cm^{-2}.s^{-1}$, 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.

As far as the spallation products are concerned, a large number of isotopes are actually contributing to the total activity. During the irradiation phase, the total activity almost saturates at 5x10^5 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 a 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.

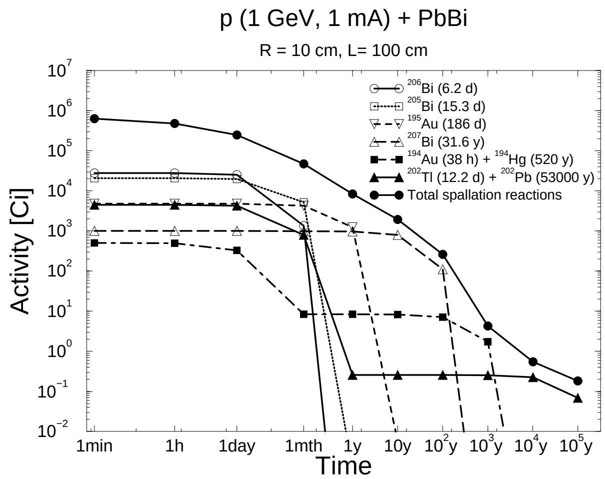

Figure 2. Activity (in Curies) of a Pb-Bi target as a function of time, after one year of irradiation, due to the spallation products (solid line), activation by the neutron flux below 20 MeV in the case of a bare target (dotted line) or in the case of a target surrounded by D_2O (dashed-dotted line).

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.

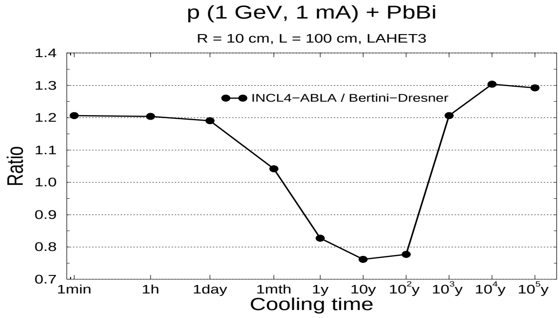

Figure 3. Ratio of the activity due to the spallation products calculated using the INCL4-ABLA and the Bertini-Dresner models, for a Pb-Bi target, as a function of time after one year of irradiation.

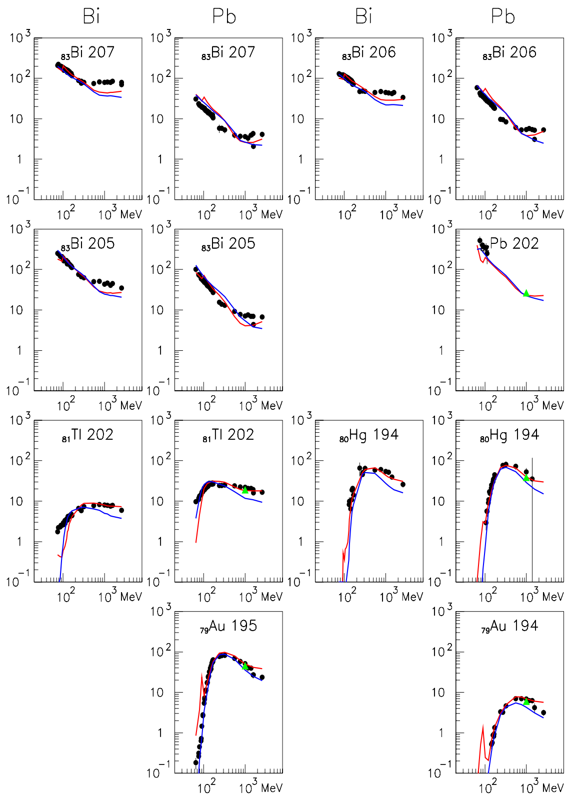

Figure 4. Comparison of INCL4-ABLA and Bertini-Dresner predictions with the production cross-sections (mb) of a few isotopes (which are the main contributors to the activity in a thick Pb-Bi target) measured in p+Pb or p+Bi by γ-spectroscopy [13] as a function of the incident energy.

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 (see fig.1), 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. As already said, the model agrees totally with the isotopic production at 1 GeV. However, in a such thick target, the primary proton induces on average one high-energy and two secondary lower energy interactions, so it is also important to predict correctly the energy dependence of the production cross-sections. This is the case, down at least to a few tens of MeV, as it can be seen in fig.4 where the elementary production cross-sections of the isotopes found to be the main contributors, measured by γ-spectrometry [13] on Pb and Bi targets, are compared to the models. The discrepancies between the model and the data being at most 30-40%, a similar uncertainty can be assessed for the predicted activity.

2.3 *Production of Impurities*

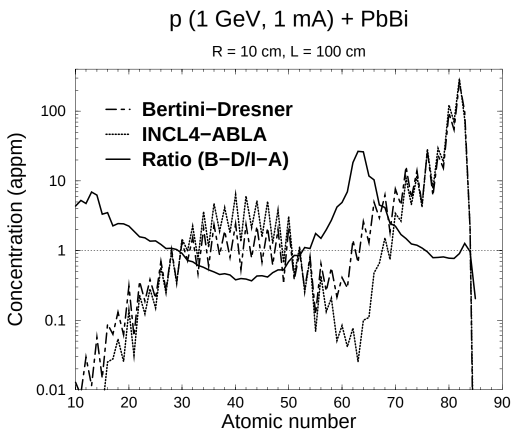

Figure 5. Concentration in appm of the different chemical impurities produced by spallation in a Pb-Bi target after one year of irradiation, calculated with LAHET3 using the INCL4-ABLA (dotted curve) or the Bertini-Dresner (dashed-dotted curve) model and ratio between the two calculations (solid line).

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. The ratio between the two calculations (solid line) is also shown. Here the discrepancy between the two models can reach a factor 3 in the region of fission fragments and up to 30 for the very light evaporation residues.

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 that INCL4-ABLA reproduces 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.

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 I-A/B-D
		Bert-Dres	Incl4-Abla	
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**

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. The element concentrations foreseen by the two models differ by a factor 2 maximum as found in fig.5 for these elements. 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.

3 Chemical impurities and DPAs in an ADS window

Spallation residues produced in p+Fe reactions at several energies (300, 500, 750, 1000 and 1500 MeV) have also been recently measured at GSI with the reverse kinematics technique [14]. The window of an ADS can be considered as a thin target and in most of the projects is foreseen to be made of martensitic steels T91 or EM10, materials containing nearly 90 % of iron. Therefore, the p+Fe isotopic cross-sections can be used directly to calculate the chemical impurity produced after a given operation time. Modifications of the chemical composition of the window material possibly result into problems of corrosion or alloy cohesion and modification of mechanical properties. Damages arise also because of atom displacements (DPA) due to spallation residue recoiling. Since the GSI experiments also allow to measure the residue longitudinal velocities, it is possible to estimate the associated DPAs. Results of impurity production and DPAs obtained from the experimental data at the five different bombarding energies have also been compared to calculations performed with LAHET3, using standard models (actually the Bertini-Dresner combination) or the new INCL4-ABLA model.

3.1 *Impurities*

A window of 2 mm thickness made of iron has been assumed to protect a cylindrical Pb-Bi spallation target surrounded by a heavy water moderator. The experimental isotopic cross-section data have used to predict the concentration of the different elements directly created by the proton beam, after one year of irradiation with a 31.8 μA/cm2 proton beam at 1 GeV. The radioactive decay of the different isotopes during the irradiation time was calculated with the ORIHET evolution code. The window is also hit by back-scattered particles coming from the spallation target and the surrounding materials. The LAHET3 code was used to simulate concentration of impurities due to these particles. Activation by the low energy neutron flux was obtained using the DARWIN code. Final concentrations are given in appm (atoms of impurities in one million atoms of the window). They are plotted in Fig. 9. The full blue line represents the total concentration as a function of the element number after one year of irradiation. The dashed blue line shows the contribution due to spallation reactions induced by the direct protons. This is by far the most important contribution except for iron isotopes. The concentration due to spallation reactions induced by light charge particles and neutrons coming from the surrounding medium is the black dotted line, while the black dashed-dotted one indicates the products created by activation by the neutrons with energies lower than 20 MeV. Non-negligible amount of phosphorus, calcium or sulfur, which can be a concern for embrittlement problems in the window material, are found.

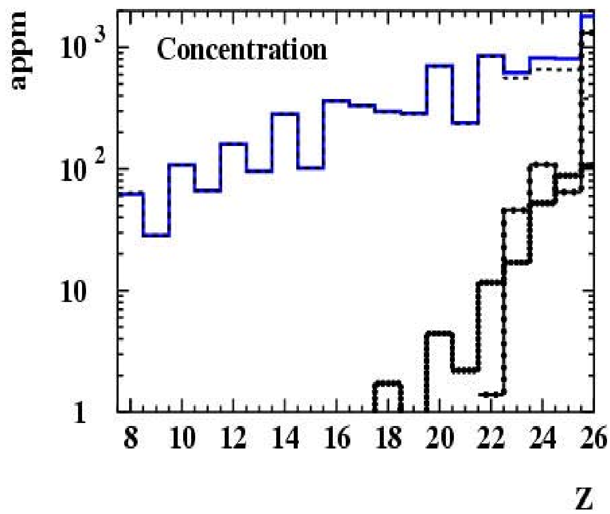

Fig. 6. Impurity concentrations versus element number in a iron window after one year of irradiation by a 1 GeV proton beam of 31.8 µA/cm2 current density. Full blue line: total concentration. Dashed blue line: concentration due to direct spallation products. Black dotted line: concentrations due to spallation reactions induced by light charge particles and neutrons coming from the surrounding medium. Black dashed-dotted line: products created by neutrons with energies lower than 20 MeV.

3.2 Damages due to recoiling residues

DPAs due to the recoiling residues have been calculated using a method based on the equation of Robinson [15] and proposed in [16]. Fig.7 shows the results obtained from the experimental cross-sections and recoil energies as a function of the incident proton energy. Also displayed are the DPAs estimated from the cross-sections and velocities predicted by the Bertini-Dresner and INCL4-ABLA. It can be seen that both models slightly underpredict the experimental results at the lowest energy. It appears that Bertini-Dresner reproduces better the recoil energies and the cross-sections for the lightest residues than INCL4-ABLA while it is the contrary for the isobaric cross-sections close to iron. Since it is the heavier fragments that most contribute to the DPAs, INCL4-ABLA is globally a little better.

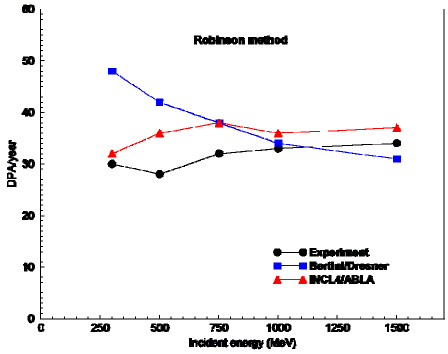

Fig. 7. Atom displacements calculated using Robinson formalism from the experimental cross-section and recoil velocities and from Bertini-Dresner and INCL4-ABLA models for the five different incident energies.

4 Conclusions

Thanks to the work performed in HINDAS, on both measurements and improvement of physics models, quantities related to the production of spallation residues in ADS can now be predicted more reliably or with a known uncertainty. Examples concerning the activity and chemical impurity production in a realistic Pb-Bi spallation target have been shown. 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. For chemical composition modifications and DPAs due to recoiling residues in the window estimation, experimental data can directly be used. However, in this case (spallation on light targets) the behaviour of the models is not yet satisfaying and work both experimentally and theoretically has still to be done.

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