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ARTICLE

Astatine production in a lead-bismuth target bombarded by a proton beam: a detailed study using INCL4.6-Abla07

Jean-Christophe David[a], Alain Boudard[a], Joseph Cugnon[b], Salim Ghali[a], Sylvie Leray[a*], Davide Mancusi[a] and Luca Zanini[c]

[a]*CEA/Saclay, IRFU/SPhN, 91191 Gif-sur-Yvette Cedex, France;* [b]*University of Liège, AGO Department, allée du 6 août 17, Bât. B5, B-4000 Liège 1, Belgium;* [c]*PSI, Zurich, Switzerland*

Liquid lead-bismuth eutectic (LBE) is often considered as a possible target in spallation neutron sources. An experiment has been performed in 2005 at ISOLDE (CERN), in which LBE has been irradiated by 1.0 and 1.4 GeV protons and isotopes of astatine, a volatile precursor of Po, have been found. Until recently no model was able to reproduce this astatine production, which is due either to double charge exchange reactions, Bi(p,π^-xn)At, or to secondary reactions induced by helium nuclei. Recently, both parts of the spallation model combination INCL4-Abla have been improved leading to the new versions, INCL4.6-ABLA07. In particular, an additional mechanism to produce light and intermediate mass fragments has been added and special care has been paid to the low energy reactions. This paper first shows that all the basic features of the different reactions leading to astatine production are well predicted by the new model. The model, implemented into a beta version of MCNPX2.7 is then used to simulate the ISOLDE experiment. A very good agreement with the experimental data is observed.

Keywords: spallation target; astatine production; spallation models

1. Introduction

Spallation targets are used in several fields: neutron sources, accelerator driven systems, radioactive ion beams, etc. Liquid lead-bismuth eutectic (LBE) is a possible choice as material for spallation targets. The spallation reactions occurring in the target give rise to a huge number of radioactive isotopes, some of which being volatile and therefore of particular concern for radioprotection. Recently, the IS419 experiment at the ISOLDE facility at CERN measured the production and release rates of volatile elements from a LBE target irradiated by a proton beam of 1 and 1.4 GeV [1]. Among others, the production of At isotopes was investigated. Although the production of astatine isotopes is relatively modest and these isotopes are generally short-lived, they could be a radioprotection issue since astatine is highly volatile and decays to polonium isotopes. In [1], the experimental results were compared to simulations with different high energy transport codes, none of which were able to predict neither the order of magnitude of the measured astatine production nor the shape of the isotopic distribution.

From the modeling point of view, the production of astatine from bismuth is interesting since its atomic charge Z=85 is larger than that of bismuth by two units. This means that it is produced through (p,π^-) reactions on bismuth or via secondary reactions involving ^{3}He or ^{4}He. Therefore, a reliable prediction of astatine yields by a given model is a test of its capability to correctly handle these particular channels.

In this paper, we use the newly developed combination of models INCL4.6 [2] and ABLA07 [3] implemented into MCNPX [4] to simulate the ISOLDE experiment. We first discuss the capability of the model to reproduce the different reaction channels involved in the production of astatine in a Pb-Bi target. Then, we present the results of the full simulation of the ISOLDE target irradiated with protons of 1.4 and 1 GeV.

2. Validation of the model

Let us recall that spallation reactions are generally viewed as a two-step process: a first stage, the intranuclear cascade, leading to the ejection of fast particles and leaving an excited remnant, which, in a second stage, de-excites through evaporation and sometimes fission. In this paper, we shall use the latest versions of the Liège intranuclear cascade model, INCL4.6, presented in details in [2], in which special attention has been paid to low incident energies and composite induced reactions and for the de-excitation, the ABLA07 model developed in GSI, Darmstadt [3].

*Corresponding author. Email: sylvie.leray@cea.fr

As already mentioned, protons irradiating a LBE target can produce astatine isotopes through the following mechanisms: ^{209}Bi(p,π^-xn)$^{210\text{-x}}$At, i.e. double charge exchange in primary reactions; secondary reactions induced by helium nuclei produced in primary collisions, ^{209}Bi(^{3}He,xn)$^{212\text{-x}}$At and ^{209}Bi(^{4}He,xn)$^{213\text{-x}}$At. Contributions from other secondary reactions have been checked to be negligible. Actually, a first simulation of the ISOLDE experiment with MCNPX has revealed that isotopes with mass larger than 209 are produced only through secondary helium-induced reactions, ^{4}He playing a larger role and leading to higher masses. On the other hand, both mechanisms populate the other isotopes, the very lightest ones preferentially originating from double charge exchange reactions.

2.1. Helium production

Since secondary reactions of helium nuclei play an important role, it is necessary to have a model able to correctly predict helium production in spallation reactions. In most models, helium is produced only in the de-excitation stage, which cannot account for the high-energy tail (above around 50 MeV) observed in the experimental spectra. Actually, only models having a specific mechanism to produce high-energy clusters of nucleons can aspire reproducing this tail. In INCL4, a mechanism based on surface coalescence in phase space has been introduced in Ref. [5]. It assumes that a cascade nucleon ready to escape at the nuclear surface can coalesce with other nucleons close enough in phase space and form a cluster that will be emitted if its energy is sufficient to overcome the Coulomb barrier.

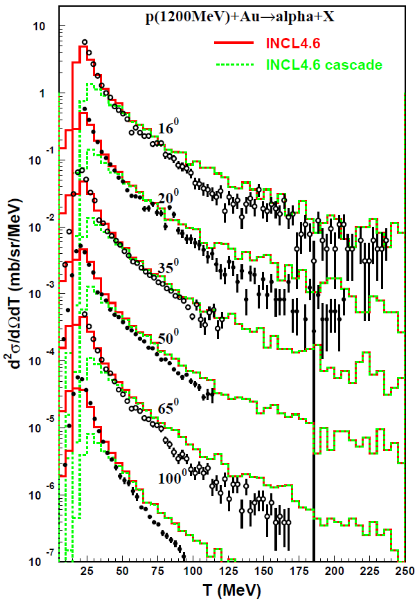

Figure 1. Alpha double-differential cross sections as a function of alpha kinetic energy, in the p+Au reaction at 1.2 GeV (from [6]) compared with the results of INCL4.6-ABLA07. The contribution due to the coalescence process in the cascade is given by the dashed green line.

In **Figure 1** experimental data from [6] of alpha production double-differential cross sections, in the p+Au reaction at 1.2 GeV, are compared to the model predictions. A very good agreement can be observed all along the energy spectrum. In order to emphasize the importance of the coalescence mechanism the dashed green curve shows its contribution in the total production cross-sections.

2.2. Helium-induced reactions

2.2.1 Total reaction cross sections

The treatment of secondary reactions induced by helium nuclei of energies below 100 MeV is also important. Although from the origin, the INCL4 model was designed to handle reactions with composite particle up to alpha, little attention had been paid to those up to recently. In addition, secondary reactions occur at low energies, generally below the alleged theoretical limit of validity of INC models. In the last version, the treatment of low-energy composite particle induced reactions has been significantly improved. Details of the modifications brought to the model are discussed in [2].

With these modifications, the model is able to predict rather well the helium-induced total reaction cross-sections as can be seen in **Figure 2** in which the model is compared to the few available experimental data [7, 8] and to the predictions of the original version.

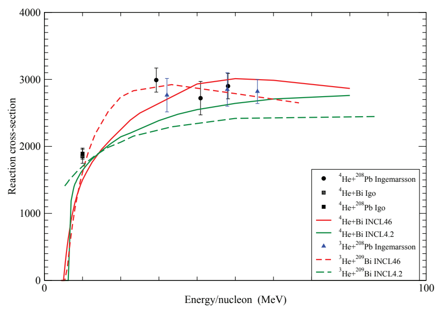

Figure 2. Total reaction cross sections as a function of incident energy for alphas (solid lines) and ^{3}He (dashed lines). on Bi calculated with INCL4.6-ABLA07 (red curves) and the older version INCL4.2-ABLA (green curves) compared to experimental data on Pb and Bi targets from [7, 8].

2.2.2 Astatine production channels

Our model correctly predicts the production of high-energy helium and the total reaction cross-sections. It remains to check that the particular (He,xn) channels involved in the prediction of the different isotopes are also under control.

In **Figure 3** experimental cross sections, found in the EXFOR experimental nuclear reaction database, regarding ^{209}Bi(α,xn), for different x values, are compared with the model. It can be observed that the model agrees rather well with the experimental results. In particular, the maximum cross-section for each channel is very well predicted. However, the opening and closing of the different channels as a function of incident energy seems to happen a little too early. This is probably related to the probability of emitting a given number of neutrons, which is very sensitive to model parameters used in the evaporation model such as the level densities or the inverse cross-sections.

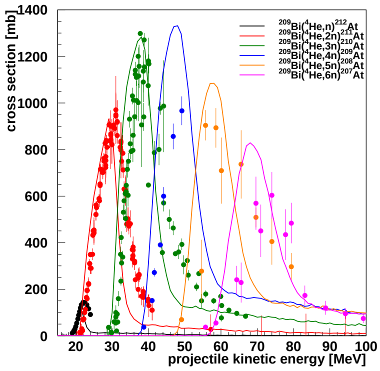

Figure 3. ^{209}Bi(α,xn) cross sections for x=1 to 6 as functions of the α incident kinetic energy. The curves correspond to the predictions of the INCL4.6+ABLA07 model. The experimental data were compiled using the experimental nuclear reaction database EXFOR.

In the case of ^{3}He, not shown here, the situation is less good. The x=1 and x=2 channels are largely overestimated, the agreement being restored only for the largest x values. However, in the ISOLDE target secondary reactions induced by ^{3}He are much less numerous than by ^{4}He for the channels with the smallest x-values. Since our model agrees well with the experimental data for ^{4}He for all x-values and for ^{3}He for x>2, we can expect the overall prediction to be reliable within a factor definitely smaller than 2.

2.3. Double charge exchange channels

Our model generally correctly predicts negative pion production. However, what matters for astatine production is the particular channel of double charge exchange, in which a negative pion is emitted without any other charged particle. The population of this channel therefore also depends upon the probability to emit protons and clusters either in the cascade stage or during the de-excitation.

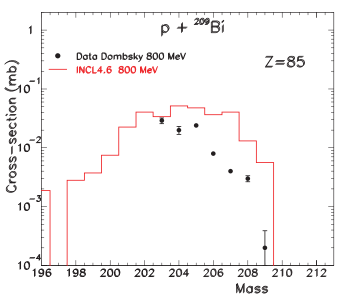

Figure 4. Cross sections for the production of different astatine isotopes from a Bi target irradiated by 800 MeV protons measured by [9] and compared to the prediction of INCL4.6-ABLA07.

Figure 4 displays the data from Dombsky et al. [9] on p+Bi at 800 MeV in which several astatine isotopes have been identified together with the result of the calculation. In both cases, the calculation overpredicts the experimental data by a factor 2 to 5 for the heaviest isotopes but less than 2 for the lightest ones. The reason for this is not well understood. It is probably linked to the ratio of charged particles versus neutrons emitted either in the evaporation or, more likely, during the cascade stage, since a larger emission of charged particle would depopulate the Z_{target}+2 production.

However, since in the ISOLDE target the double charge exchange channel is a major contribution only for light isotopes, it can be expected that our simulation for those isotopes should be correct within a factor 2 with a tendency to an overprediction.

3. Astatine production yields in the ISOLDE target

Figure 5 shows the result of the MCNPX simulation with INCL4.6-ABLA07 compared to the ISOLDE data at 1 GeV for the total production yields of astatine isotopes. A time of the order of 20 hours has been assumed for the complete release from the liquid metal during which the radioactive decay of the different isotopes is taken into account. A remarkable agreement between the calculation and the experiment is observed, regarding not only the shape of the isotopic distribution but also the absolute release rates. Clearly all the new features discussed in the preceding sections, in particular the better handling of low energy helium-induced reactions, have considerably improved the predictive capability of our model compared to the version used in [1].

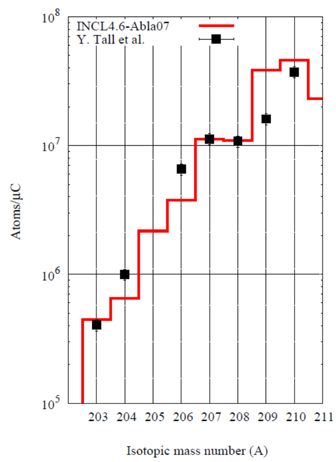

Figure 5. Astatine release rates measured by Tall et al. [1] at 1 GeV compared to MCNPX simulations, using INCL4.6-ABLA07, assuming a time of the order of 20 hours for the complete release from the liquid metal, during which the radioactive decay of the different isotopes is taken into account.

In **Figure 6**, the 1.4 GeV data are presented. In order to emphasize the importance of the secondary reactions induced by the clusters produced during the cascade stage through our coalescence mechanism, a calculation has been performed switching off this mechanism. The result is presented as the green curve and exhibits a severe deficit of heavy isotopes. Obviously, a model unable to emit high energy helium nuclei cannot be expected to correctly predict astatine production in a LBE target, since only a small fraction of the heliums produced in the evaporation stage have enough energy to undergo a reaction before being stopped. This was the case of our first version INCL4.2, which is more or less mimicked by INCL4.6 without clusters, but also of the MCNPX default model option, Bertini-Dresner.

In the same Figure, the results are also compared with CEM03 [10], which is also available in MCNPX2.7b (grey line), using the same assumption on the release time. It is interesting to note that this model is not able to account for the measured yield of the heavy astatine isotopes. In fact, CEM03 does have mechanisms to produce high-energy heliums but does not produce isotopes with mass larger that 209 probably because of the treatment of low energy helium induced reactions.

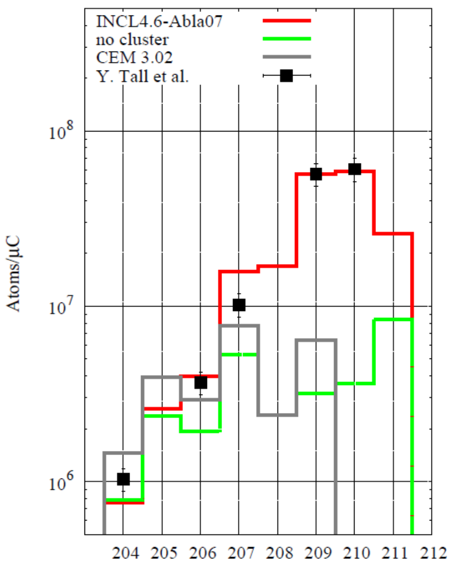

Figure 6. Same as Figure 5 but at 1.4 GeV and with additional comparisons with a calculation in which the production of clusters in the INC model was switched off (green line) and to CEM03 (grey line).

4. Conclusion

In this paper, we have investigated the capability of the new INCL4.6 version of the Liège intranuclear cascade model, coupled to the deexcitation code ABLA07, to predict astatine production in LBE spallation targets, when implemented into a high-energy transport code such as MCNPX, and applied it to the experimental data from the ISOLDE collaboration that could not be, up to now, reproduced by the different codes that were tried.

We first checked that the model reasonably reproduces all the features of the elementary interactions that play a role in the generation of isotopes with a charge larger than that of the target by two units, i.e. (p,π^-) reactions, the energy spectrum of helium nuclei produced in primary interactions and the helium-induced secondary reactions.

The full simulation of the ISOLDE experiment has been performed, assuming a time of the order of 20 hours for the complete release from the liquid metal, during which the radioactive decay of the different isotopes is taken into account. The shape of the isotopic distribution is then perfectly reproduced but also the absolute release rates. A simulation done switching off the cluster production through the coalescence mechanism in INCL4.6 stressed its importance for the astatine production. This points out that models not having a similar mechanism, as the default Bertini-Dresner model in MCNPX, cannot be expected to reliably predict astatine in LBE. CEM03, probably because of the treatment of secondary helium-induced reactions, largely underestimates the heavy isotopes.

More generally, it can be claimed that our model, thanks to the attention paid to the emission of high-energy clusters and to low-energy cluster induced reactions, can be safely used within MCNPX to calculate the production of isotopes due to secondary reactions.

Acknowledgements

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