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Nuclear Science

Utilisation and Reliability of High Power Proton Accelerators (HPPA5)

Workshop Proceedings
Mol, Belgium
6-9 May 2007

NEA No. 6259

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

APPLICATION OF THE INCL+ABLA REACTION MODEL TO THE STUDY OF THE EVOLUTION OF SPALLATION TARGETS

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Abstract

The design of spallation targets of accelerator-driven systems requires the use of reliable spallation reaction models embedded inside general transport codes. Nuclear reactions above ~150 MeV are often reasonably well described by a Monte Carlo implementation of an intranuclear cascade (INC) model followed by a de-excitation model. The Liège intranuclear cascade model (INCL4) coupled to the ABLA evaporation-fission model has been shown to give a fairly good description of a large amount of experimental measurements for proton-induced spallation reactions on thin targets in the 200 MeV-2 GeV range, without free parameters. This model was recently improved in order to extend its capabilities and to cure some remaining shortcomings. These modifications bear on nuclear mean field, some aspects of pion dynamics, light charged clusters emission and low-energy extensions. In this paper we will compare both the modified and the standard versions of the INCL model with respect to their ability to reproduce thin target experimental measurements. After, we will tentatively validate the improved INCL model for particle emission and for residue production in thick targets. In order to estimate the residue production, MCNPX has been updated and coupled to the ORIGEN evolution code, through the use of the ALEPH system developed at SCK•CEN. The constraints on the reaction models brought by the thick target data, especially concerning radiotoxicity, will be compared with those provided by the similar comparison with thin target data.

Introduction

The projects of accelerator-driven systems for the transmutation of nuclear waste have increased the interest in spallation reactions as primary neutron sources. The design of such facilities requires the use of reliable spallation reaction models, embedded inside general transport codes, to accurately evaluate emitted particle distributions, radiation damages and radiotoxicity of the target.

A spallation reaction is currently described by a first brief phase of intranuclear cascade process governed by nucleon-nucleon collisions, leading to an excited nucleus after ejection of a few energetic particles, followed by a second longer phase corresponding to the decay by evaporation with a possible competition with fission.

The intranuclear cascade model of Liège INCL4 [1] coupled to the ABLA evaporation-fission model [2] has been shown to give a fairly good description of a large amount of experimental measurements for proton-induced spallation reactions on thin targets in the 200 MeV-2 GeV range [1]. These last years this model was improved to extend its capabilities and to cure some remaining shortcomings [3]. The INCL4 model and these recent extensions will be shortly presented in the next section.

After comparing the standard and the modified versions concerning their predictions for particle emission and residue production in thin targets, validating the modified model, we will investigate whether these extensions have an influence on particle emission and on the residue production inside a thick target. This validation will allow studying the impact of spallation model improvements on particle transport inside media.

The INCL4 model

The standard version of the Liège INC model is described in detail in Ref. [1]. It is important to recall that the particle-nucleus collision is described by a Monte Carlo simulation. Initially the position and momentum of all target nucleons are randomly distributed in the nuclear volume and in a Fermi sphere, respectively. In the INCL4 version, a smooth initial density distribution is introduced, in concordance with electron scattering data. $NN \Leftrightarrow NN$, $NN \Leftrightarrow N\Delta$ and $\Delta \Leftrightarrow \pi N$ collisions are based on realistic parameterised cross-sections and are subjected to a consistent Pauli blocking. The cascade is stopped according to a criterion based on the time evolution of some physical quantities. The code accommodates nucleons and light composites as incident particles.

This leads to a parameter-free code in the 200 MeV-2 GeV range with its own absolute normalisation (the computed total reaction cross-section is correctly reproduced). Neutron and proton energy spectra are well reproduced for a vast set of target nuclei and incident energies (see Figures 1 and 2). Concerning residue production, the rates are on average well reproduced, except for the isotopes containing more protons than the target nucleus, which are overestimated (see Figures 3 and 4) and for the rare earths which are underestimated (not shown here).

The recent extensions of the INCL42 model consist of:

- the introduction of an isospin and energy-dependent baryon mean field [4];
- an improvement of the pion dynamics: new cross-sections and introduction of the pion mean field [5];
- several refinements allowing the extension to lower energy [6-8];

- the implementation of d, t, ^{3}He and ^{4}He production during the intranuclear cascade step [9];
- extension to higher energy, up to 20 GeV [10].

Since the last two extensions are still in progress, they will not be considered in our studies below. Despite the first reasonably good results obtained for incident energies as low as a few tens of MeV, among the low-energy extensions, only the refinement of the Pauli blocking for the first collision will be used in our studies. Some of these low-energy ingredients can not be easily taken into account at the same time as the energy dependence of the nucleon potential.

Thin target results

The modification of the baryon mean field leads to an improvement (reduction) of the multiplicity of neutrons per cascade. The excitation energy of the nucleus after the INC step and the multiplicity of neutrons per evaporation are increased. Figure 1 gives the double-differential cross-section for neutron production induced by 800-MeV protons on ^{208}Pb. Globally, the effects of our modifications are rather small: a slight decrease of the intensity of the quasi-elastic peak and a shift towards lower energy are observed, the production of intermediate-energy neutrons is slightly reduced and the emission of evaporated neutrons is increased. Compared to the experimental measurements, our modified model improves the capabilities of INCL. Since the quasi-elastic peak, and thus events involving few emitted particles, is improved by our modifications, we also expect an improvement in the production of residues close to the target isotope (see below).

Figure 1. Double-differential cross-section of production of neutrons induced by 800-MeV protons on ^{208}Pb

A zoom of the quasi-elastic peak is given at the top of the figure. The results obtained with the standard and the modified models are given by the dotted line and the continuous line, respectively. Experimental data come from Ref. [11].

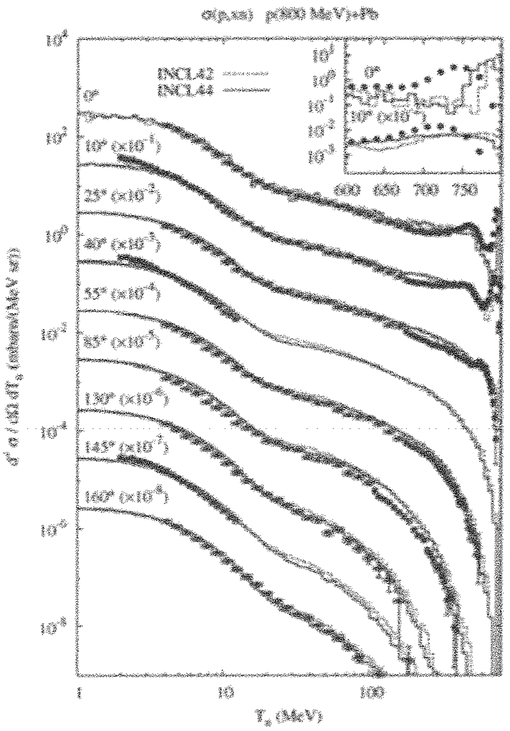
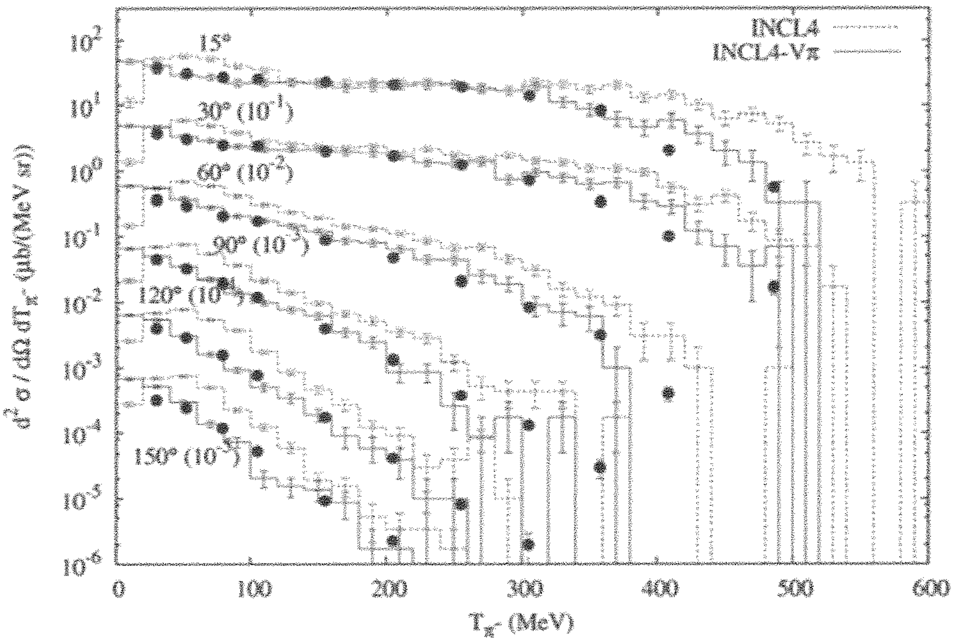


Figure 2 gives the double-differential cross-section for π^- production induced by 730-MeV protons on ^{208}Pb. This figure shows that the production of pions is considerably improved by our modifications. This will also have an impact on the production of some residues: above 100 MeV, the only way to produce ^{210}Po from ^{209}Bi is the (p,π^0) channel. The globally good agreement seems to show that the most important characteristics of the pion and Δ dynamics are taken into account in the modified INCL model.

Figure 2. Double-differential cross-section for production of negative pions induced by 730-MeV protons on ^{208}Pb

Same convention as in Figure 1. Experimental data are taken from Ref. [12].



Figures 3 and 4 give the excitation function for the production of ^{210}Po and ^{209}Po induced by protons on a ^{209}Bi nucleus. As expected from the improvement of the estimation of the quasi-elastic peak and of the pion sector, the production of these elements is improved by our modifications. It should be noted that concerning the production of residues, the overall good agreement observed with the standard model remains and the increase of the excitation energy after the cascade step does not degrade the fission residue production but slightly improves the production of rare-earth isotopes (not shown here, see Ref. [5]).

Thick target results

We have investigated whether the previous improvements survive for a thick target. To this aim, we have updated the INCL model implemented in the MCNPX transport code [16]. Figure 5 displays the double-differential multiplicity of neutrons emitted from a thick Pb target (60 cm length and 10 cm radius) irradiated at the Saturne laboratory by 800-MeV protons [17]. Globally, our modifications bring smaller effects. This can be explained by compensation effects: less primary neutrons but of higher energy will produce more neutrons in secondary collisions.

Figure 3. Excitation of the cross-section for ^{210}Po production induced by protons on ^{209}Bi

Same convention as in Figure 1. Experimental data are taken from Ref. [13].

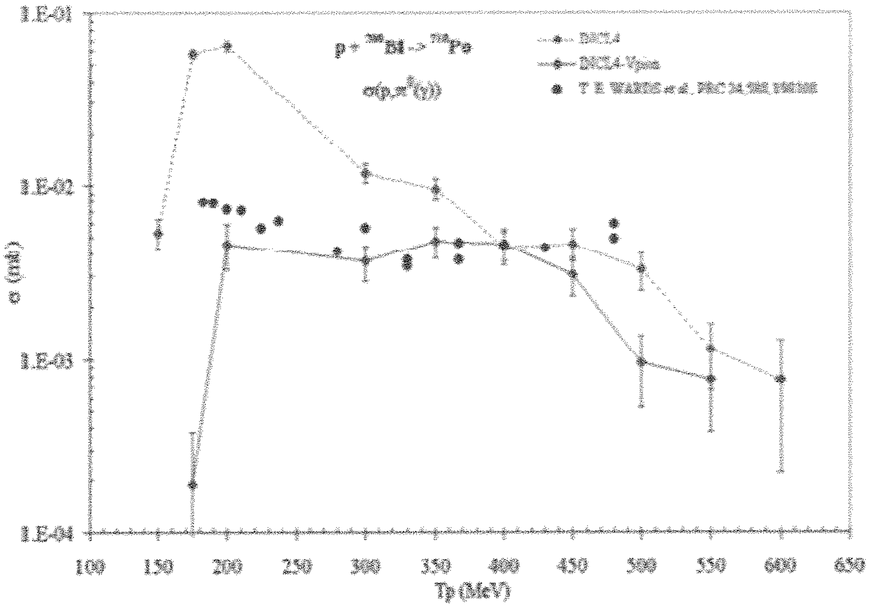


Figure 4. Excitation function of the cross-section for ^{210}Po production induced by protons on ^{209}Bi

Same convention as in Figure 1. Experimental data are taken from Refs. [14,15].

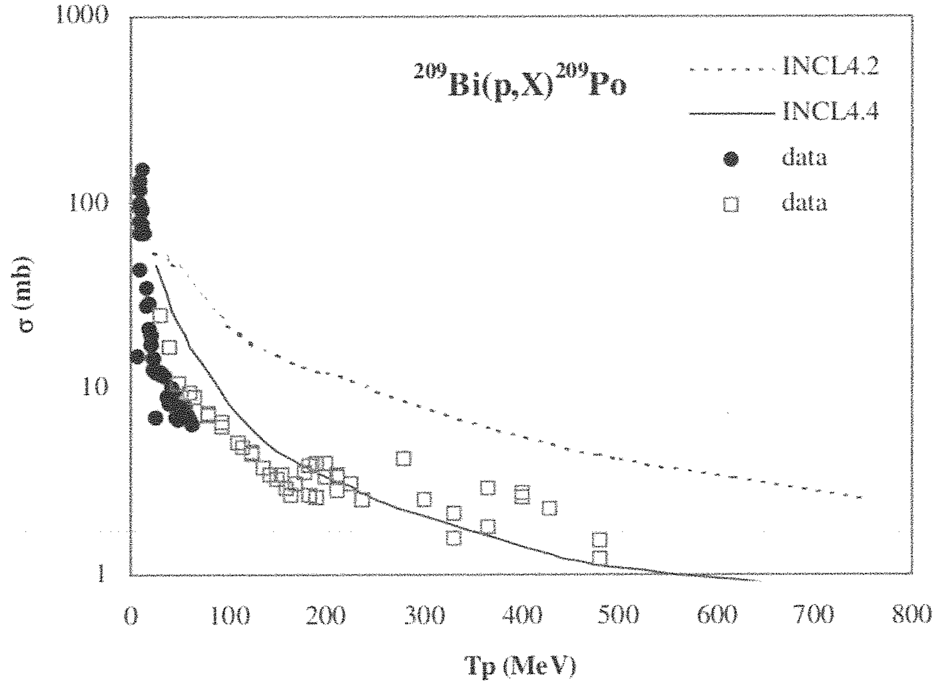
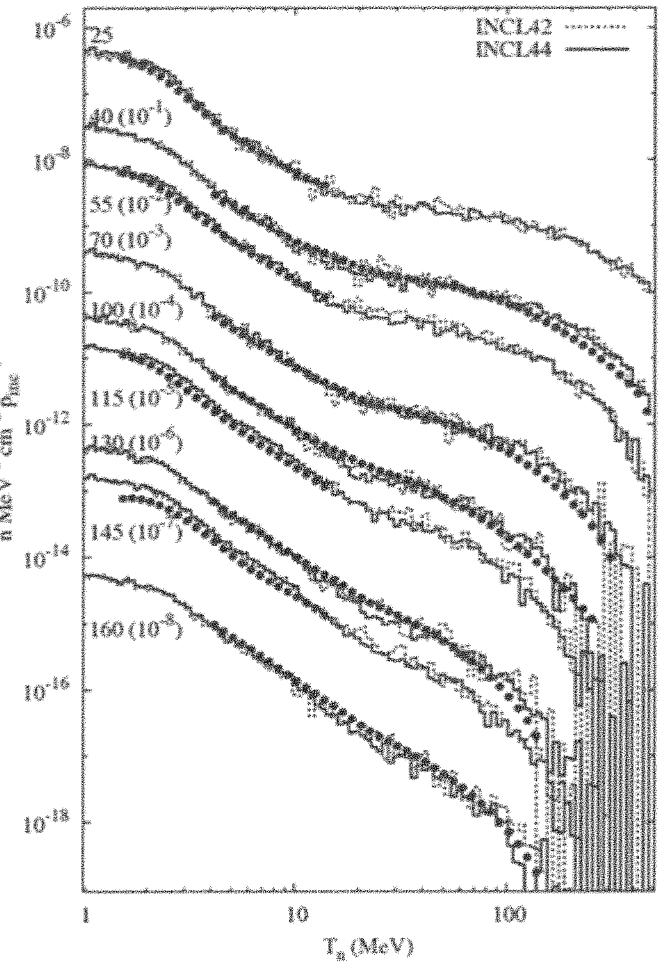


Figure 5. Double-differential multiplicity of neutrons induced by protons of 800 MeV on a thick Pb target (see text for detail)

Same convention as in Figure 1. Experimental values are taken from Ref. [17].



We will now investigate whether the improvement of the production of residues remains in the thick target case. In order to estimate the evolution of the spallation target residues we have solved the multi-particle Bateman equations:

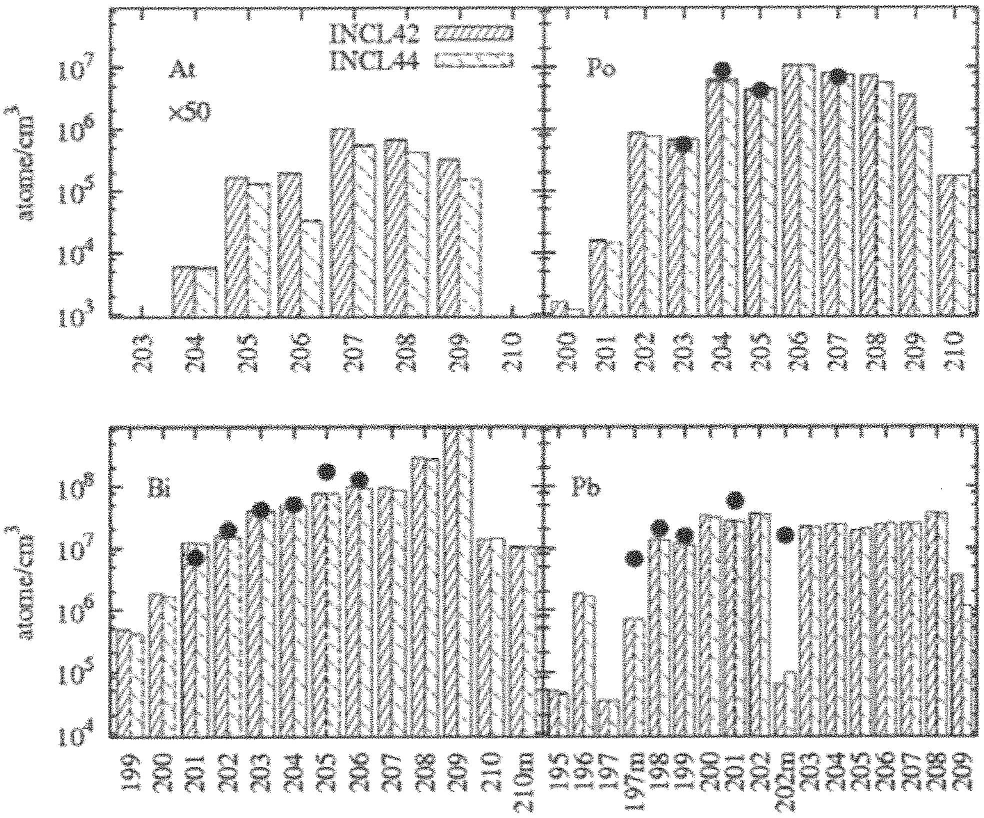
$$\frac{dN_i}{dt} = \sum_{j \neq i} \sum_{k=n,p,\pi} \alpha_{j,k,i} \sigma_{j,k} \phi_k N_j + \sum_{j \neq i} \beta_{j,i} \lambda_j N_j - \sum_{k=n,p,\pi} \sigma_{i,k} \phi_k N_i - \lambda_i N_i$$

using the ALEPH [18] code which couples the MCNPX code and the evolution code ORIGEN [19]. As ALEPH was initially developed for "low" energy (E < 20 MeV) neutrons, we have adapted this code to accommodate the evolution of spallation residues [20]. Residue production induced by protons and higher energy neutrons' interaction are extracted from MCNPX using the HISTP output file and the HTAPE3X post-processing tool. The decay library of ALPEH is also updated using NUBASE97 [21].

The experimental measurements used to validate the production of residues inside a thick target are taken from Ref. [22]. In this experiment, a stack of alternating Pb and Bi disks was shortly irradiated by 590-MeV protons at PSI and the residues were measured by γ-spectrometry. In Figure 6

Figure 6. Isotopic density of At, Po, Bi and Pb produced in the second disk (^{209}Bi)

Experimental data are taken from Ref. [24]. Figure adapted from Ref. [25].



the theoretical estimates obtained in the first Bi disk are confronted to the experimental results of Ref. [23]. Both the standard and the modified INCL models reproduce the experimental results. The production of isomeric nuclei is strongly underestimated. This is due to the use of old data for the residual γ de-excitation (PHTLIB data file) provided with MCNPX version 26a [24]. As foreseen from thin target analyses, ^{208}Po and ^{209}Po are reduced with the same magnitude. The production of ^{210}Po is not influenced by our modifications and more ^{210}Po are produced inside a thick target than a thin one. In a thick target, the (n,γ) reaction is of course the dominant source of production of ^{210}Po.

Only the first eleven disks were analysed in Ref. [23]. The calculated production of the most highly radiotoxic isotopes is given in Figure 7 for the Bi disks, which contain most of the radiotoxic isotopes [25]. Except for the production of ^{210}Po, which is fed by the low-energy neutron capture reactions, the production of the other isotopes remains quite constant inside the successive disks and the reduction of Po isotopes is basically constant inside these disks.

Figure 8 displays the calculated evolution of some of the most radiotoxic isotopes produced in the LBE target of the MYRRHA ADS [26]. The irradiation consists of 1.88 mA and 600-MeV protons during 270 EFPDs. After this irradiation, the target decays during 1 000 years. We can draw the same conclusion as for the PSI target except that here the production rates of ^{210m}Bi and of ^{210}Po are still larger due to the contribution of the neutrons coming from the surrounding subcritical core which enhances the (n,γ) reaction rate on ^{209}Bi.

Figure 7. Activity (in Bq/cm^3) of ^{204}Bi and ^{194}Hg induced in the Pb disks

Same convention as in Figure 1. Figure adapted from Ref. [25].

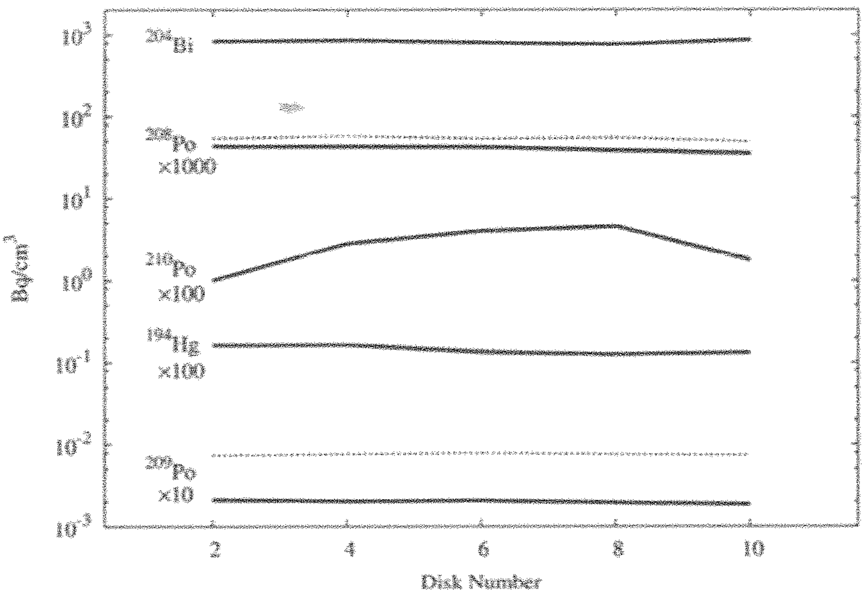
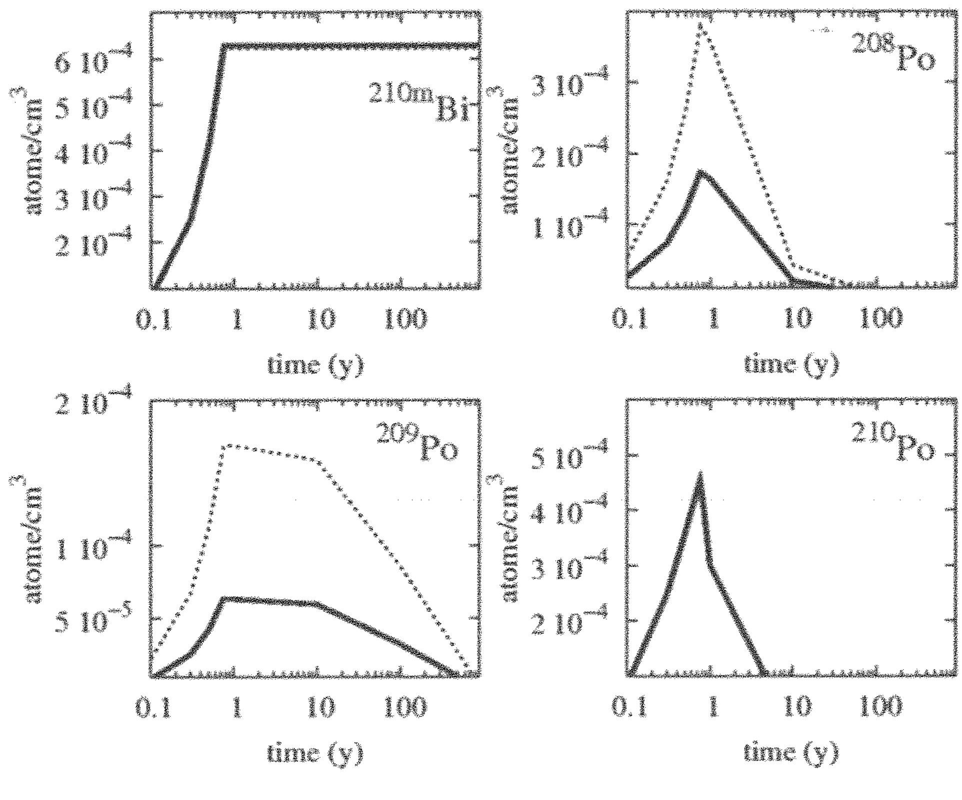


Figure 8. Evolution of some of the most radiotoxic isotopes produced in the LBE target of the MYRRHA ADS

Same convention as in Figure 1



Conclusion

Extensions of the INCL model allowed a better description of events involving the emission of few particles and, consequently, the production of polonium residues induced by protons in thin ^{209}Bi targets. The predicted ^{210}Po and of ^{209}Po production cross-sections are considerably reduced and are now very close to the experimental measurements.

To assess the influence of spallation models' improvements on thick spallation target design, the INCL model implemented in the MCNPX code was updated. If the impact of our modifications on the emission of nucleons is reduced by compensation effects inside thick targets, for the production of residues, the variation observed is of the same magnitude for thin and thick targets.

The effect of our improvements for the production of some radiotoxic isotopes induced in thick targets were first performed for the stack of alternating Pb and Bi disks bombarded with 590-MeV protons at PSI. The evolution of this spallation target was performed by adapting the ALEPH code. For the few experimental measurements, the predictions of the standard and the modified INCL models remain close to each other and give satisfactory results. The use of old data for the residual γ de-excitation in MCNPX26a gives a strong underestimate of the production of isomeric states. As observed from our thin target analysis, the production of ^{209}Po and ^{208}Po is strongly reduced. Comparing the effects of our modifications in thin and thick targets, the observed variations due to our modifications remain similar. The production of ^{210}Po and of ^{210m}Bi in the bismuth disks are not influenced by our new implementations, since these isotopes are mainly produced by (n,γ) reactions, followed, for the ^{210}Po, by β^- decay.

In the case of the LBE target of the MYRRHA ADS, the same conclusion as for the PSI target can be drawn, except for of ^{210}Po and of ^{210m}Bi for which the production rates are larger due to the presence of the surrounding multiplying media.

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