MEGAPIE: RESIDUE YIELDS AND RADIOACTIVITY PREDICTIONS WITH DIFFERENT MODELS IN MCNPX

J.-C. David, A. Boudard, S. Lemaire, S. Leray, S. Panebianco
CEA Saclay, DSM/DAPNIA, France

Abstract

During the last years, numerous experiments dedicated to spallation reactions have been performed and coupled to the development of codes. Among these studies, a lot of efforts have been devoted to the validation of the intra-nuclear cascade (INC) model, INCL4 [1], developed in a collaboration between Saclay and the University of Liège, combined with the evaporation-fission model, Abla [2] from GSI. These models have been implemented into the transport code MCNPX2.5.0 [3], so that we can easily compare them to the other models already included (Bertini [4] and Isabel [5] for INC part, Dresner [6] for the deexcitation step, and also the CEM2k [7] stand alone combination). In this paper, we will study the residue production in a *real* spallation target, the Megapie target [8] that will be irradiated next July at PSI. The differences between the models predictions for masses and activity rates will be shown and discussed.

Introduction

Spallation reactions, and their by-products, appear as a useful tool for basic research, technological applications and even mankind purposes. Some of the nuclei produced in these reactions are exotic, neutron rich for example. Thus numerous facilities, referred as Radioactive Ion Beam (RIB), already built (Spiral - GANIL), planned (Spiral2-GANIL, Fair-GSI) or studied (Eurisol, RIA) are based on these reactions. Another important feature is the large amount of emitted neutrons during the process (20-25 neutrons per proton for the reaction p(1GeV)+Pb, for instance). These neutrons can be used to drive subcritical reactors, so called Accelerator Driven System (ADS), which could be helpful for nuclear waste transmutation. Some projects already exist such as Myrrha or SAD. These neutrons can also directly irradiate materials as it is done in material testing reactors, but with a larger spectrum. The American Spallation Neutron Source (SNS) which started this year is one of the existing examples.

Spallation reactions consist of an energetic light particle impinging a nucleus. Particle can be a nucleon or a light ion (d, t, ^{3}He, α) with a kinetic energy ranging from 100-200 MeV.A to about 2.5 GeV.A. Some models are able to give unexpected rather good results even below 100 MeV [9]. Due to the large range of energy considered we can not rely on data as it is done for low energy neutrons (below 20 or 150 MeV), and models are needed. Those models consider a reaction often divided into two steps: a first and fast one, the IntraNuclear Cascade (INC), followed by the second slower, the deexcitation (evaporation and/or fission). A third one is sometimes proposed in between, the preequilibrium stage.

We can find numerous models for the INC (Bertini, Isabel, INCL4, CEM, FLUKA (peanut) [10] …) and for the deexcitation as well (Dresner (with RAL [11] or ORNL [12] for fission), GEM [13], Abla, FLUKA …). Last years several of these models have been improved thanks to the new data obtained for light particle production (mainly neutrons, but also light charged particles) and for residue production (through inverse kinematics (GSI) or γ-spectrometry). Data mentioned here are devoted to thin targets, where models can be easily tested. Existing data on thick targets are scarce and principally focused on neutron production. To reproduce them we first have to develop models that produce the correct level and nature of emitted particles, with their spectra. They must also describe the reaction for incident particle in a wide energy range, because of secondary particle emission, with lower energies than the primary projectile and inducing also spallation reactions.

At the CEA-Saclay, our spallation group is involved, on the experimental side, with measurements at Saturne [14] (particle) and GSI (residue within FRS [15] and now Spaladin [16]), but also, on the modelling side, with INCL4, and finally in the implementation of INCL4 and Abla in the transport codes LAHET3.16 [17] and MCNPX2.5.0. Comparisons of models to data have been done within different frameworks (Hindas [18], Eurisol [19]…). We are now interested in doing some predictions for real spallation targets and especially Megapie. We will present in the following comparisons of different codes or code combinations available in MCNPX to experimental data (thin and thick targets). Then we will explain how we can obtain, with the use of CINDER'90 combined with MCNPX, the masses of the residues and activities associated for a given irradiation time and at several steps after shutdown. Finally, we will show the results obtained with different models in MCNPX for the Megapie target.

Models/data comparisons

The use of tools can only be efficient if one knows how good they are for a given purpose.

Comparison of models to data for thin target (test of the physics of the model for given projectile, energy and target) and for thick targets (real targets, test of all possible incident particles with a wide energy range) is a tremendous task. So we will show, in the following, few results obtained with the different models included in MCNPX and will compare them to experimental data.

Thin targets

Data shown or discussed below are taken from [20] for neutrons, [21] for the light charged particles and [15], [22] for the residues.

Neutron

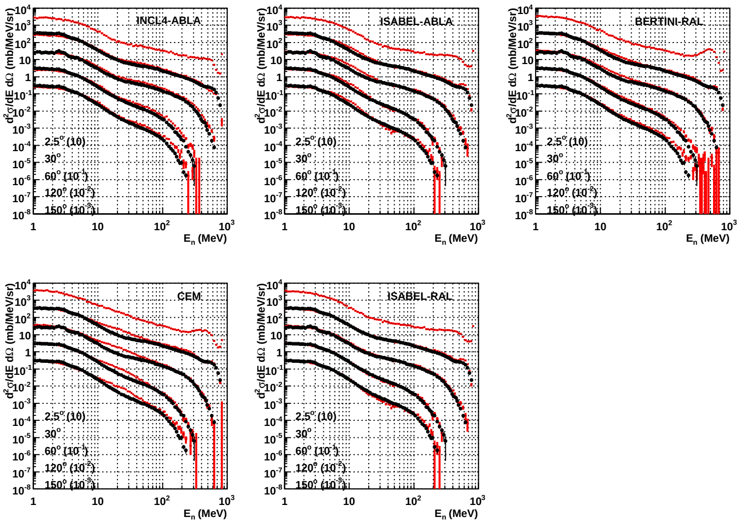


Figure 1: Neutron spectra for the reaction p(800MeV)+Pb – five model combinations available in MCNPX2.5.0 (here “RAL” means Dresner combined with RAL fission model)

It has to be mentioned that Bertini is always used with the preequilibrium process, as recommended by the authors of LAHET3.16.

Neutron energy spectrum is easily reproduced by the different models. Their agreement with experimental data is quite good (fig 1), even if some improvements remain to be done.

Light charged particle

Figures discussed below can be seen in the B. Rapp et al. report “BENCHMARKING OF THE MODELLING TOOLS WITHIN THE EURISOL DS PROJECT”, task 4 of this SATIF-8 meeting.

Proton spectra are not so well reproduced as for neutrons, but results are still good.

The situation is different for α spectra. Whatever the model used the result is in strong disagreement with experimental data. If we focussed our attention on INCL4-Abla, the shortcomings are due to the fact INCL4 doesn’t emit α (or any other lcp with A>1), so it misses the high energy part of the energy spectrum. In addition Abla, for the evaporation stage, do not consider the emission of d, t or 3He and parameters or ingredients, used to model the evaporation process, are sometimes too simple (coulomb barriers and capture cross sections, for instance). New improved versions of INCL4 and Abla solving these problems are studied and available, but not yet implemented in transport codes.

Residue

INCL4-Abla (or Isabel-Abla) reproduces well the mass and charge distributions, except for the residues produced after a long evaporation process. Bertini-Dresner reaches the same level of agreement except for the fission part (fig 2) which is in bad agreement with experimental data.

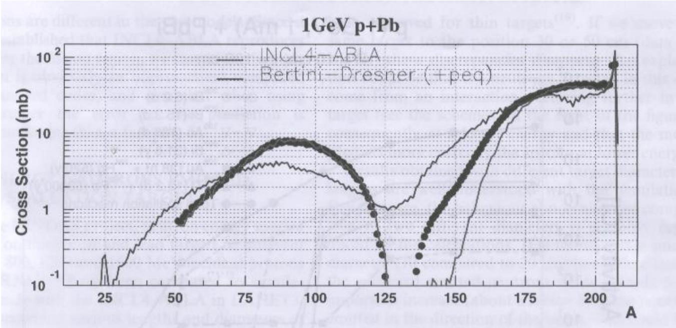


Figure 2: Mass distribution for p+Pb at 1 GeV.A

INCL4-Abla is able to reproduce quite well excitation functions (fig 3), which is not always the case for Bertini-Dresner.

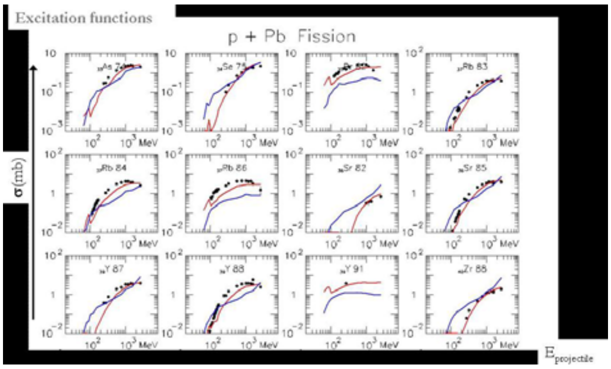


Figure 3: Excitation functions for the reaction p(70MeV→2.6GeV)+Pb
INCL4_Abla is red and Bertini_Dresner blue

Thick targets

Data are scarce compared to thin targets and mainly dedicated to neutron spectra.
References used below are [23] for the neutrons and [24] for the residues.

Neutron

As for thin targets, the neutron spectra are equally well reproduced by the models. As a result the choice of the model is not crucial for such observables.

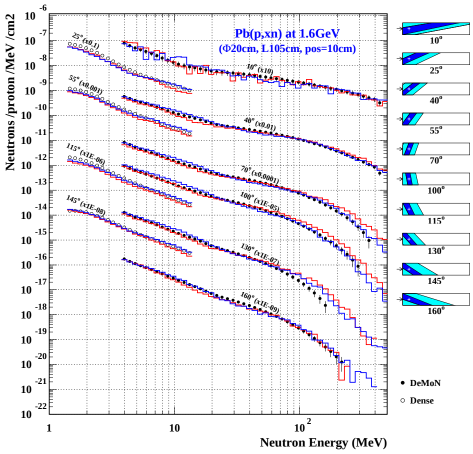


Figure 4: Comparisons of neutron spectra between SATURNE data (black points), INCL4_Abla model (red line) and Bertini_Dresner model (blue line) for p(1.6GeV)+Pb.

Residue

Very few data exist for residue production in thick target. Here we show Xe isotopic distributions obtained at CERN with an UCx target. Discrepancies appear between the models predictions. INCL4-Abla and Isabel-Abla give rather good results.

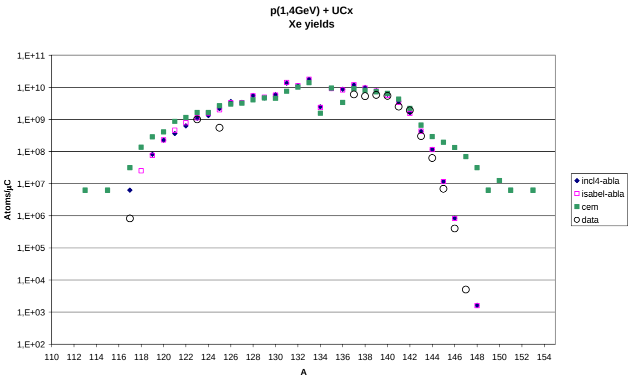


Figure 5: Production yields of Xe isotopes from ISOLDE UC_x targets. Three models are compared to the data.

MCNPX-CINDER’90

For each spallation targets, there are two different steps: irradiation and cooling time. In order to calculate nuclei production one needs to produce the nuclei and let them decay, since most of them are radioactive. The production is due to the spallation reaction, but also to neutron reactions at low energies. Then, to get the nuclei produced at a given time (during irradiation and after) we use for spallation residue production the multi-particle transport code MCNPX2.5.0 combined with the material evolution program CINDER’90 [25]. It will compute the nuclei produced by low energy neutrons (spectrum given by MCNPX) and take into account the radioactive decays.

Megapie

The Megapie project, at PSI (Switzerland) within the SINQ facility, aims at demonstrating the feasibility of a liquid Lead-Bismuth spallation target [8]. The beam will be a 575 MeV and 1.4 mA proton beam (~0.8 MW). A detailed view is shown below (fig 6).

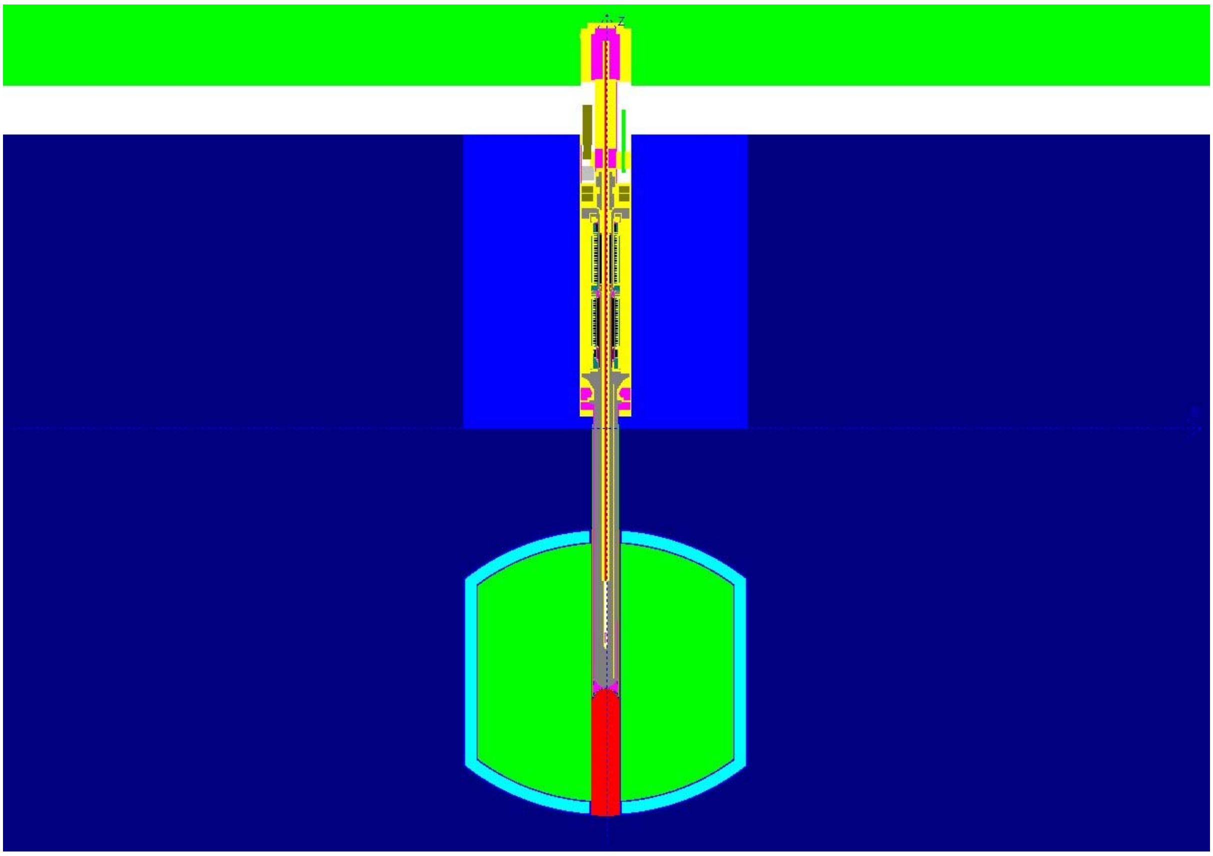


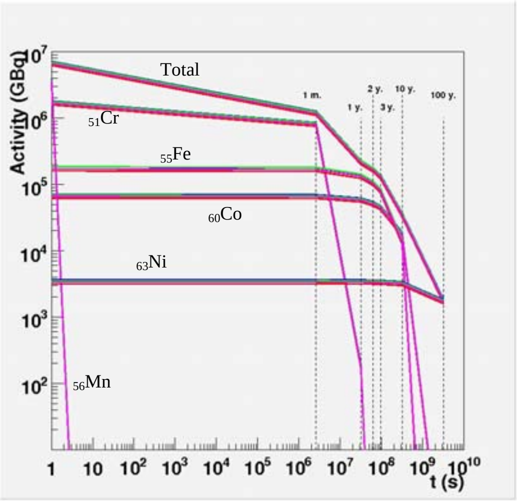
Figure 6: Megapie geometry used for MCNPX.
Proton beam comes from the bottom. The liquid Pb-Bi target (grey) is surrounded by heavy water (green). The upper part is dedicated to the cooling.

A comprehensive work has already been performed within the X9 group for the R&D of Megapie [26]. Proton and neutron fluxes, power deposition, radiation damage and isotope production have been calculated with several code systems. For all of these observables no significant differences are found, except for isotope production. Since for this benchmark MCNPX was only used with the default option for the high energy part, that is Bertini for the INC stage and Dresner for the deexcitation stage, we decided to compare this default option to the other better and/or still improved combination models, which are INCL4-Abla, Isabel-Abla and CEM2k.

Activities in four different places

Activities obtained by the different codes are compared in four different places made of four different materials. These materials are 316L (Stainless steel), T91 (mainly Iron), AlMg3 and Pb-Bi target. 316L is in the central rod (yellow in figure 6). T91 is around the Pb-Bi target from the bottom to the top. AlMg3 is between the heavy water (green) and the light water (blue).

316L



INCL4-Abla
Isabel-Abla
CEM2k
Bertini-Dresner

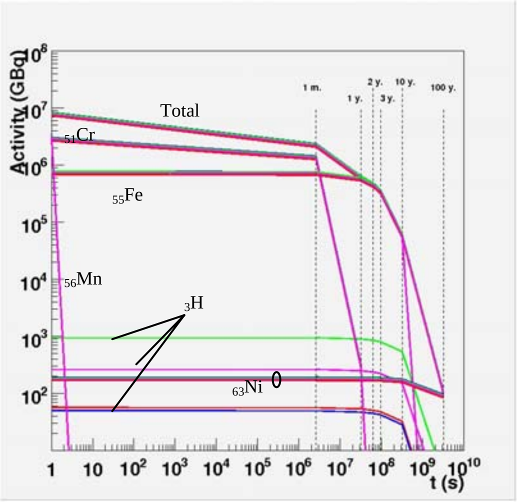
	1s	1m	1y	2y	3y	10y	100y
Mn56	15 6 12	15 6 12					
Cr51	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12		
Fe55	15 7 16	15 7 16	15 7 16	15 7 16	15 7 16	15 7 16	15 7 16
Co60	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12
Ni63	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12

Isabel_Abla
Bertini-Dresner
CEM2k

Figure 7: On the left, activities are plotted for above mentioned models. Total and main contributors are shown. On the right a table gives the ratio (%) to INCL4_Abla for main contributors

The models give the same results within around 15 %.

T91



INCL4-Abla
Isabel-Abla
CEM2k
Bertini-Dresner

	1s	1m	1y	2y	3y	10y	100y
H 3	-12 366 1578	-12 366 1578	-12 366 1578	-12 366 1578	-12 366 1578	-12 366 1578	-12 366 1578
Cr51	15 6 12	15 6 12	15 6 12	15 6 12	15 6 12		
Mn56	15 6 12	15 6 12					
Fe55	14 7 14	14 7 14	14 7 14	14 7 14	14 7 14	14 7 14	14 7 14
Ni63	15 6 11	15 6 11	15 6 11	15 6 11	15 6 11	15 6 11	15 6 11

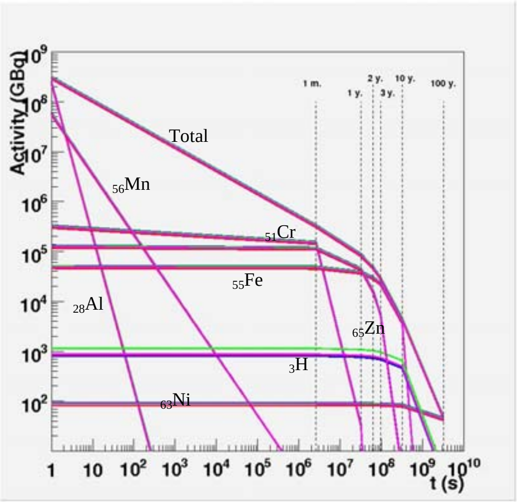
Isabel_Abla
Bertini-Dresner
CEM2k

Figure 8: See figure 7 for explanations

All results agree within 15 %, except for the activity coming from ^{3}H where Isabel and INCL4 give the same value, while Bertini and CEM2k are respectively 4 times and 16 times higher. This isotope comes from either the spallation reaction or from low energy neutron reaction. In fact this result is not surprising since neither INCL4, Isabel nor Abla emits tritium when Bertini-Dresner and CEM2k do it. Since we saw that these models produce approximately the same neutron flux (fig 1), the low energy neutron interactions can not explain the production of ^{3}H. The implementation of tritium emission by INCL4 and Abla is in progress, and we know that the production of tritium by

CEM2k is too high as it can be seen on the figure 5 of the B. Rapp et al. report, task 4 of this SATIF-8 meeting.

AlMg3



INCL4-Abla
Isabel-Abla
CEM2k
Bertini-Dresner

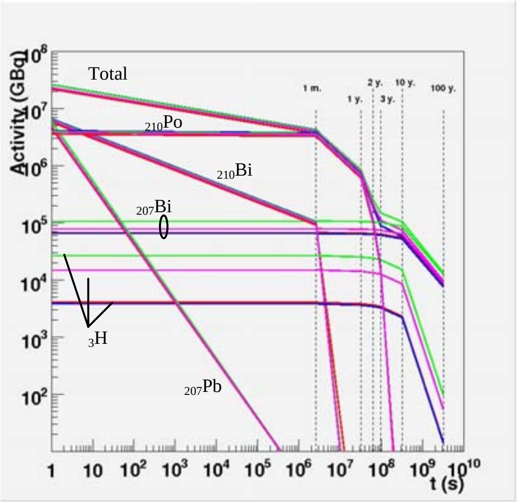
	1s	1m	1y	2y	3y	10y	100y
H 3	-5 4 36	-5 4 36	-5 4 36	-5 4 36	-5 4 36	-5 4 36	-5 4 36
Al28	13 6 11	-11 5 32					
Cr51	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11		
Mn56	13 6 11						
Fe55	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11
Ni63	14 6 11	14 6 11	14 6 11	14 6 11	14 6 11	14 6 11	14 6 11
Zn65	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11	13 6 11	

Isabel_Abla
Bertini-Dresner
CEM2k

Figure 9: See figure 7 for explanations

The conclusions are similar to the previous ones for T91, except that here the material is far from the beam. As a consequence, only the low energy neutrons play a role for the tritium production. That explains the little difference observed (36 %).

Pb-Bi target



INCL4-Abla
Isabel-Abla
CEM2k
Bertini-Dresner

	1s	1m	1y	2y	3y	10y	100y
H3	-6 268 557	-6 268 557	-6 268 557	-6 268 557	-6 268 557	-6 268 557	-6 268 557
Pb207	28 8 29						
Bi207	-4 16 59	-4 16 59	-4 16 59	-4 16 59	-4 16 59	-4 16 59	-4 16 59
Bi210	15 7 11	15 7 11	-92 -92 -89	-92 -92 -89	-92 -92 -89	-92 -92 -89	-92 -92 -89
Po210	15 7 12	15 7 11	15 7 11	15 7 11	15 7 11	-36 -40 -36	-92 -92 -89

Isabel_Abla
Bertini-Dresner
CEM2k

Figure 10: See figure 7 for explanations

The situation is a bit more complicated in the target. All models agree within 30 % or even less, except for tritium, ^{207}Bi and ^{210}Bi. The ^{3}H case has been explained previously. For ^{207}Bi the difference is not so important since only CEM2k show a discrepancy around 60%. To explain it, one should first know from which channel this isotope is produce, ^{206}Pb is a candidate, and secondly, look at the behaviour of these models on experimental isotopic distributions for element close to the target. Concerning ^{210}Bi differences begin to appear only after 1 year. This isotope can be produced directly by spallation, but also by the radioactive decay of ^{210}Pb, and INCL4 produces more ^{210}Pb than all other models.

Volatiles

We plot below H and Xe masses and activities ratio to INCL4_Abla for the three others models: Isabel_Abla, CEM2k and Bertini_Dresner.

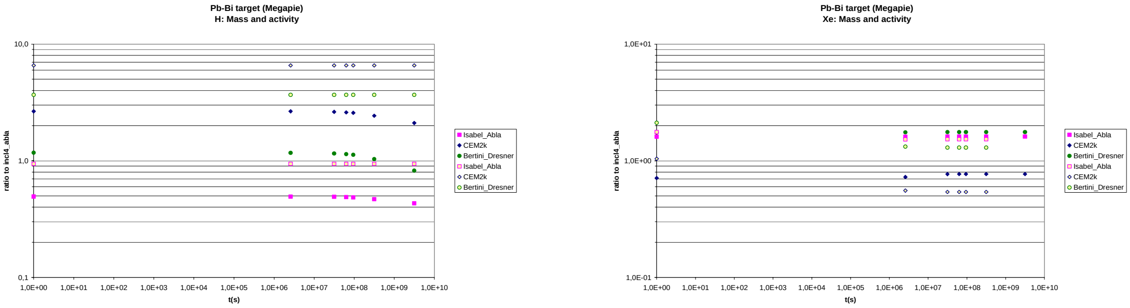


Figure 11: H (left) and Xe (right) activity and mass (bi-colour points) ratio to INCL4_Abla for Isabel_Abla, CEM2k and Bertini_Dresner model

For Hydrogen, mass is mainly due to proton and activity to tritium, so we observe the same differences or similarities between the models as the ones seen before concerning tritium.

For Xenon, coming from the fission process, the results are within a factor two. If mass and activity have different ratios, the reason has to be found in the isotopic distributions. Thus Zanini et al. [27] showed the same behaviour for Bertini_Dresner compared to INCL4_Abla, since the former predicts bigger mass than the latter. Nevertheless, one have to mention that in this paper results are obtained with a 1.4 GeV proton beam while the Megapie beam is a 0.575 GeV proton beam, so, if it gives us a explanation, energies and materials considered must be as close as possible and one have to use it carefully. For example, if we change the material, UCx instead of Pb-Bi, with a 1.4 GeV proton beam, the ratio of Isabel_Abla obtained here (fig 11) is not easy to explain when looking at fig. 5.

Conclusion

This study aimed at comparing models available in the transport code MCNPX2.5.0: default option Bertini-Dresner and three other models, some still under development, INCL4-Abla, Isabel-Abla and CEM2k. Comparisons have been performed for the Megapie project that is a liquid lead-bismuth spallation target.

The results obtained on activities, with four different materials, but also on masses for volatiles elements in the target, show some discrepancies. Different reasons may explain these results. Fission process modelling can explain the differences for volatile like Xe (figure 11). For tritium production, some models have to improve the evaporation predictions. Finally, the INC models may give a different level of isotope production for nuclei close to the spallation target (^{210}Bi).

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