NUDATRA – WP 5.4 – Task T5.4.3

Activation calculations for the MEGAPIE target with INCL4 and ABLA, and comparison with other codes

J.-C. David, S. Lemaire and S. Leray*

CEA/Saclay, IRFU/SPhN, 91191 Gif-sur-Yvette Cedex, France

1. Introduction

The final objective of the WP5.4 workpackage of NUDATRA is to provide nuclear models in the 200-2000 MeV energy range for implementation into high-energy transport codes, so that most quantities related to high-energy reactions, required for the design of future ADS, can be calculated reliably or at least with a known uncertainty. In this domain, the elementary interaction is generally described as a two-step mechanism by an intranuclear cascade model followed by a de-excitation stage. In the former FP5 HINDAS project [1], an important effort had been devoted to the collection of high quality experimental data and, simultaneously, to the development of improved spallation models. As a result of this work, the combination of the intranuclear cascade model, INCL4 [2], and de-excitation model, ABLA [3], has been implemented into MCNPX [4], after having been tested against available data and shown to give globally better predictions than models used by default (Bertini-Dresner [5]). Spallation models have also been benchmarked for other applications. As an example, for the EURISOL-DS project, several models have been tested with a proton beam of about 1 GeV hitting different materials. Particle and residual nucleus yields were calculated [6] with different models available in MCNPX2.5.0, which are generally the combination of an intra-nuclear cascade code followed by a de-excitation stage: INCL4-ABLA, ISABEL [7] coupled to ABLA, Bertini-Dresner and CEM2k† [8]. In all cases, important remaining deficiencies were identified, concerning principally light evaporation residue and light charged particle production predictions. The solving of these deficiencies, which should lead to new improved versions of INCL4 and ABLA, is one of the goals of WP5.4. Task T5.4.3 is devoted to the validation of the new models in ADS target simulations. Calculations of the total activity and volatile element production in a real spallation target, actually the MEGAPIE target [9] recently irradiated at PSI, have been performed with the version of INCL4-ABLA currently available in MCNPX, and compared with other model calculations in order to point out possible differences due to the choice of physics models. In a second step, similar calculations with the newly developed versions of INCL4 and ABLA will be done to assess the impact of the improvements made in WP5.4.

MEGAPIE (MEGAwatt PIlot Experiment) is a liquid lead-bismuth eutectic (LBE) spallation target built to be tested with a beam power approaching 1 MW at the SINQ continuous spallation neutron source at PSI, Zürich. Neutrons produced in the target are slowed down to thermal energies in a heavy-water tank (Fig. 1). The MEGAPIE target was installed in place of the usual solid target. The

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* Present address: CEA/Bruyères-le-Châtel, DAM Ile de France, 91297 Arpajon Cedex, France

† CEM2k in MCNPX2.5.0 is a stand-alone model where the cascade exciton model CEM2k is combined with a deexcitation model.

LBE (82 litres) is circulating in a vertical loop over a length of about 5 meters. The lower part of the LBE container (red part in Fig.1 Left), called the container in the following, is made of T91 steel. A double wall, D_2O cooled, hull made of $AlMg_3$ ensures the separation from the beam line vacuum. In our calculations, we call window the lower part of the T91 container (green part in Fig.1 Left). The 575 MeV proton beam, hitting the target from the bottom, was delivered with an average current of 0.947 mA during 123 days. The reader could find more details in [10].

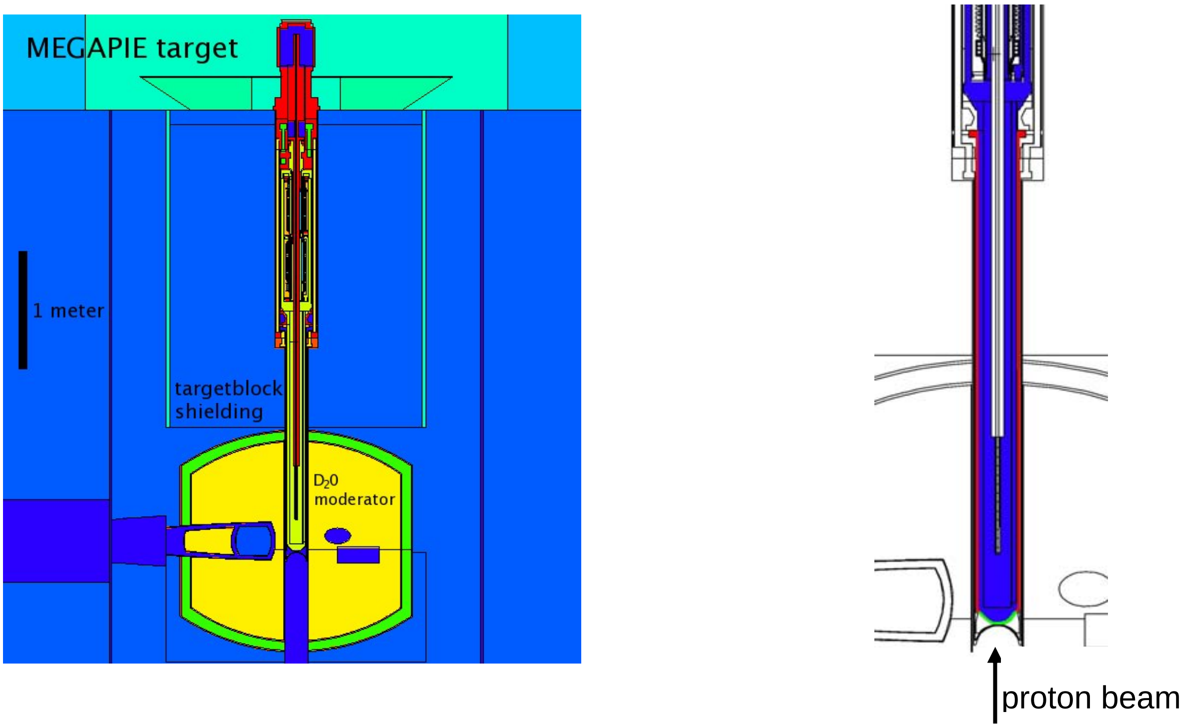

Figure 1: MCNPX 2D visualization of the simulated system. Left: the whole SINQ facility. Right: the lower part of the MEGAPIE target. The LBE is colored in blue, the container in red and green and its bottom part, the window, in green. Figures are drawn from [10]

After having explained the methodology used we present activities in the target, in the container and in the window, calculated by four models available in MCNPX2.5.0. The role of the low energy neutrons (E < 20 MeV) is studied as well as the contribution of the main radioisotopes to the activity. In order to better understand the results and to study the structure changes, mass and charge distributions are shown. Finally, we discuss the volatile element (in particular noble gas) production and the effects of impurities in the material composition.

2. Methodology

Nuclear transport code packages are generally used and coupled to evolution codes to simulate the functioning of a spallation target. Transport codes usually consist of the coupling of a high-energy part relying on spallation models and a low energy part utilizing nuclear data libraries (for neutron-induced reactions below 20 MeV). In our paper, the transport code MCNPX2.5.0 is used to simulate the full geometry of the MEGAPIE target inside the SINQ facility. Calculations have been performed with four different combinations of intranuclear cascade and de-excitation models available in MCNPX: Bertini-Dresner (default option), ISABEL-ABLA, INCL4-ABLA and CEM2k. The evolution code CINDER'90 [11] was combined to MCNPX to take into account the subsequent decay of the spallation products and activation by the low energy neutron flux. Details of this methodology are given in a schematic view in Fig. 2.

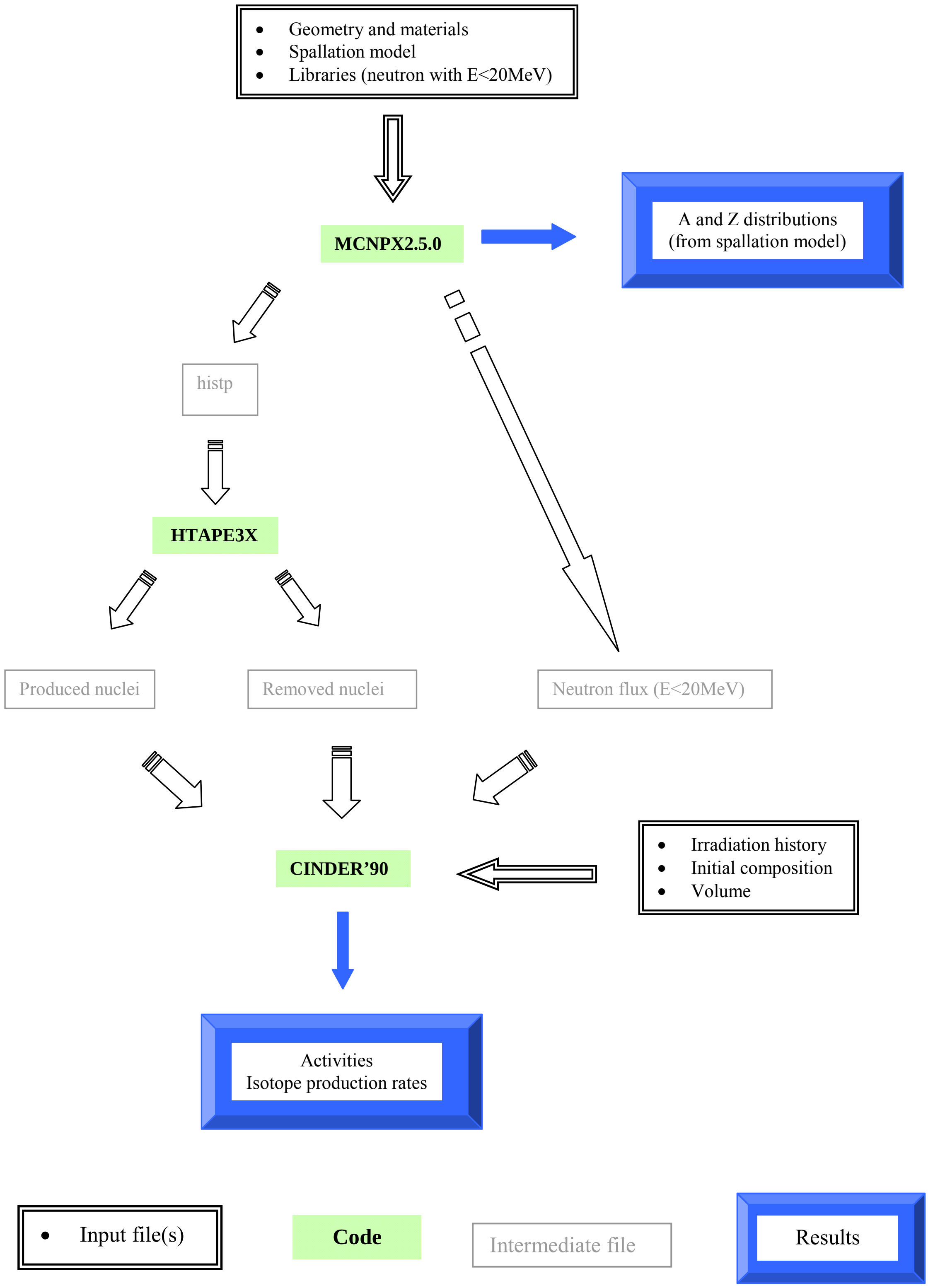

Figure 2: Schematic view of the calculation methodology

3. Results

3.1. Total activity calculations

3.1.1. Activity of the different component of the target

The evolution with time of the total activity of the different parts of the target (LBE, container and window) has been calculated until 100000 years after the shutdown. On Fig. 3, only the results obtained with the INCL4-ABLA model are plotted (see next section for model comparison). In each case, the solid line stands for the total activity while the dashed line shows the contribution due to residual nuclei produced in high-energy (spallation) reactions and calculated with the models. Actually, proton induced reactions are calculated at all energies with the models. The difference between the two curves corresponds to the activation by the low energy (E<20 MeV) neutron flux.

As expected, the highest value of activity is found in the LBE, around 10^{16} Bq at the end of the irradiation. This result can be explained by the fact that most of the incident protons interact inside the LBE creating secondary particles, part of which also interacts in the LBE. In fact, the specific activity (Bq/g) is reduced by the dilution due to the circulating loop of LBE, 82 litres, 850 kg, to be compared to the volume of the lower container, ~5 litres, 40 kg. The activity decreases rather slowly with time, remaining at a level of 10^9 Bq after 10^5 years, mainly because of the production of long-lived residues (see below for the discussion on the main contributors). The comparison between the solid and the dashed curves show that the activity is dominantly produced in spallation reactions. Only around 100 days is the contribution of activation by the low energy neutron flux visible. This is due to neutron capture by ^{209}Bi leading to ^{210}Po (half-life: 138 days).

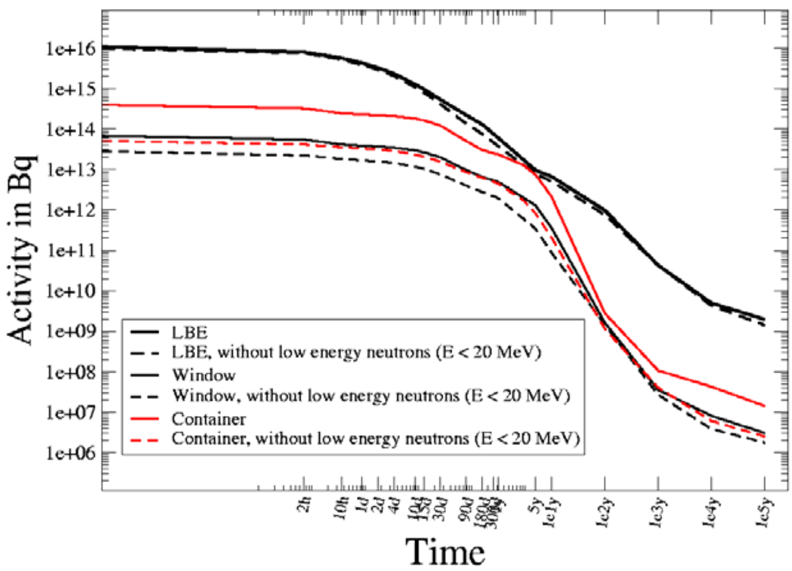

Figure 3: Activity of the LBE, container and window. Full activities are in solid lines and activities without low energy neutron (E < 20 MeV) contribution are in dashed lines. The calculation is done using INCL4-ABLA.

In the container and in the window the total activity is lower than in the LBE but the specific activities are higher, especially in the window (~0.1 litre, 800 g), which is directly hit by the proton beam. After the shutdown, the activity in the both parts decreases more rapidly than in the LBE, in particular between 5 and 1000 years. Contrarily to the LBE, the contribution of the low energy neutrons (E < 20 MeV) is dominating the activity in the container, which sees only secondary neutrons and the flux of neutrons slowed down in the heavy water. In the window, about half of the activity is due to activation. This could seem surprising since the window is directly exposed to the beam but it should be reminded that the mean free path of a 600 MeV proton is of the order of 15 cm. This means that only a small part of the proton beam interact inside the window.

3.1.2. Comparison of results obtain with different models

As mentioned previously we have compared four spallation models available in MCNPX2.5.0, Bertini-Dresner being the default option generally used in spallation target simulations. Fig. 4 shows the comparison for the LBE. As already found in previous works [12] at higher incident energies, all models give more or less the same total activities except around 10 years, which corresponds to the half-life of tritium. Around this time, tritium is indeed one of the major contributors in the case of Bertini-Dresner (blue curve) and CEM2k (green curve) while it is little produced in the case INCL4-ABLA and ISABEL-ABLA. This is illustrated by the blue and red dashed curve, which represents the contribution of tritium in respectively the Bertini-Dresner and INCL4-ABLA calculations. In fact, the tritium shown by the red dashed curve comes only from (n,t) reactions induced by neutrons between 10 and 20 MeV in CINDER. The absence of tritium production is actually a well-known deficiency of the ABLA model, in which only neutrons, protons and alphas are considered in the evaporation process. This means that calculations of tritium production should not be done using ABLA as the de-excitation code. This problem will be solved in the new version of ABLA developed in WP5.4. Also, the production of high-energy tritium isotopes observed in elementary experimental data but not predicted by any model up to now will be explicitly treated in the new version of INCL4 [13]. Even if the results concerning tritium production can be considered as more reliable with CEM2k and Bertini-Dresner, it should be stressed that these models were found in [6] to be discrepant by a factor 2 to 3 when compared to elementary experimental data. This gives an idea of the degree of predictability of the simulation.

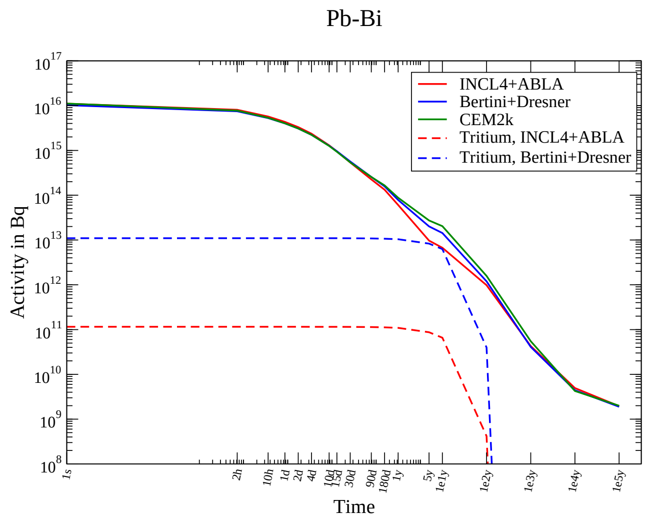

Figure 4: Total activity of the LBE as a function of cooling time after 123 days of irradiation with 575 MeV protons at 0.947 mA using different models. Dashed curves show the contribution of tritium to the total activity.

Concerning the container, the results (not shown here) even more similar with the different models than for the LBE. This is not surprising since the activity is mostly due to the activation by the low energy neutron flux, which is found nearly identical with the different models. Actually, neutron production in spallation reactions has been extensively validated both on elementary data and on simple thick target configurations [14, 15]: generally, all the models agree with the data within 10 to 20%. This means that, as long as the activation cross-sections are well established, calculations of the activation by the neutron flux can be considered as reliable. For the window, see below.

3.2. Main contributors to the activity

In order to be more precise, the main contributors to the activity in the three parts of the target have been studied and are displayed in Figs. 5 to 7 for calculations with INCL4-ABLA and Bertini-Dresner. In the LBE (Fig. 5), the activity is produced by a large number of different isotopes. For instance, there is no radionuclide contributing more than 5% up to 10 days after the end of irradiation. This reflects the fact that the activity is mainly produced in spallation reactions, which generate a wide variety of residues, as seen in elementary reactions [16, 17]. The production cross-sections being larger for nuclei close to the target materials and for neutron-poor isotopes, the main contributors appearing along the time are β^+-emitters close to Pb and Bi. As already said, the only important contributor coming from activation is the ^{210}Po around 3 months. The production of isotopes close to the target nucleus involves reactions in which only a few nucleon-nucleon collisions occur during the intranuclear cascade stage and lead to low excitation energies. The fact that calculations with different models give nearly identical activity distributions, except in the already discussed case of tritium, is therefore not surprising since actually all models describe similarly this type of reactions, discrepancies appearing for lighter isotope production. Furthermore, it has been shown that, for these isotopes, the models, in particular INCL4-ABLA, are in very good agreement with elementary isotopic distributions measured in Pb at 0.5 [17] and 1 GeV [16] at GSI, and excitation functions measured for Pb and Bi. This means that the calculations presented here can be safely relied on.

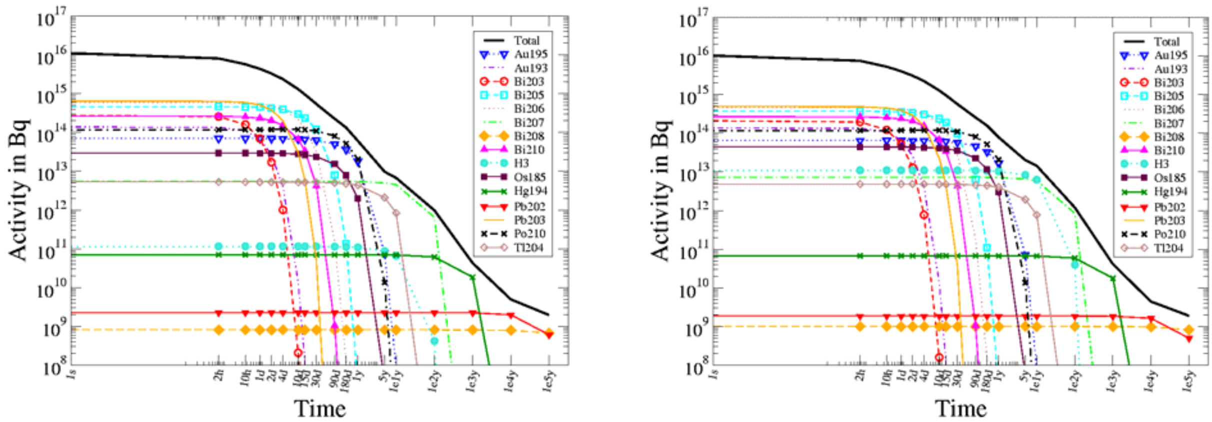

Figure 5: Main contributors to the activity of the LBE, as a function of cooling time after 123 days of irradiation with 575 MeV protons at 0.947 mA using INCL4-ABLA (left panel) and Bertini-Dresner (right panel) models.

In the container (Fig. 6), the activity is shared only among a few isotopes, resulting from the activation by the low-energy neutron flux of the components of the T91. As time evolves, the major contributor is successively: ^{51}Cr, ^{55}Fe, ^{63}Ni and ^{59}Ni. In the window, the situation is a little different since the activity is due half to activation and half to spallation reactions. This can be seen in Fig. 7, which shows that, in addition, new isotopes formed by spallation, as ^{39}Ar, ^{44}Sc and ^{44}Ti play a role in particular around 100 years after irradiation. In the case of ^{44}Ti, it can be observed that the predictions by the two models differ substantially. This reflects differences actually observed in the predictions of elementary production cross-sections in p+^{56}Fe for isotopes relatively far from the target nucleus of the reaction. The case of ^{39}Ar, a volatile isotope, will be discussed later.

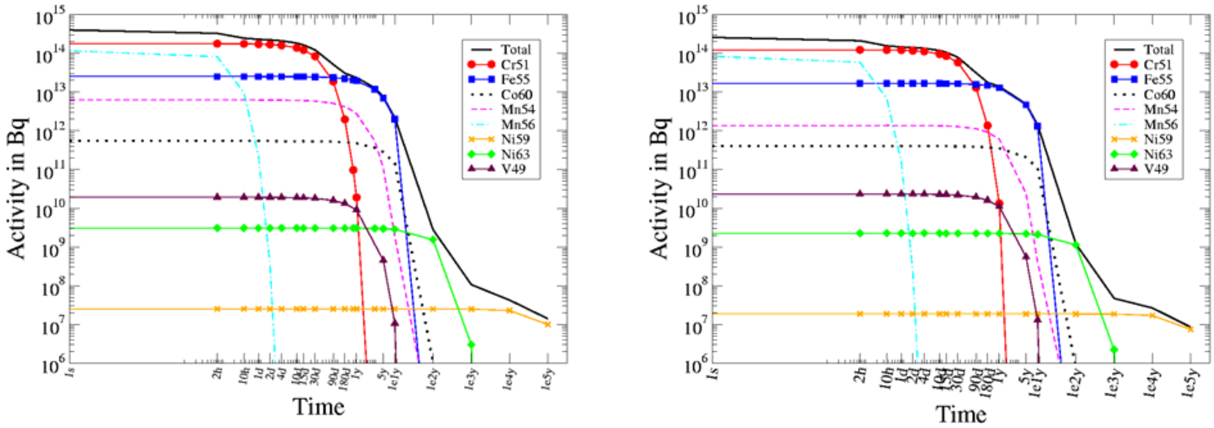

Figure 6: Same as Fig. 5 for the container.

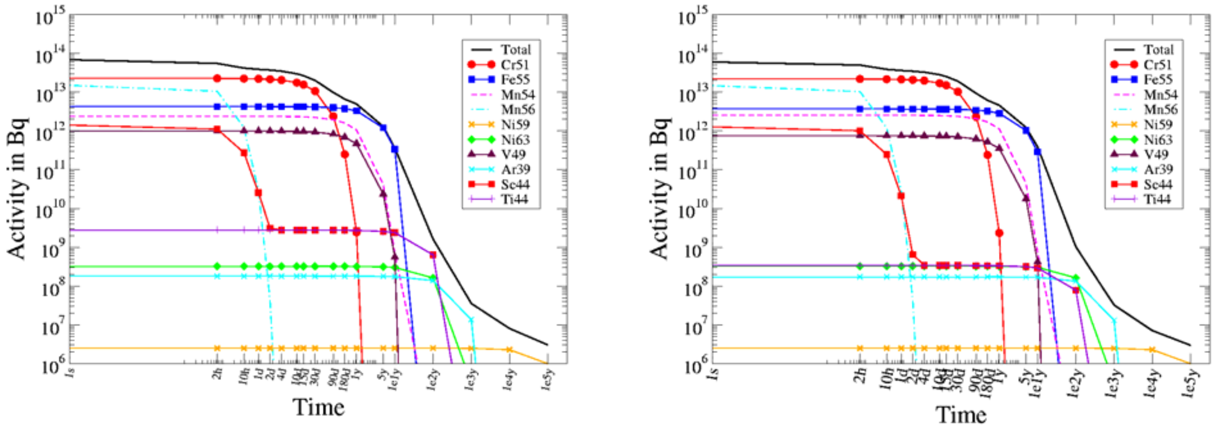

Figure 7: Same as Fig. 5 for the window.

3.3. Volatile elements

Some of the radionuclides produced in the target are volatile elements. Although their quantity is too low to play a significant role in the total activity of the target, one has to pay attention to them, because they are continuously during operation and are potentially dangerous in case of a containment failure. Therefore, the volumes of the gases produced in the LBE (Fig. 8) and their activities (Fig. 11) must be known. The mechanisms producing the different volatile elements are different. H and He are evaporated particles, Kr, Xe and I are fission products, Ne and Ar elements may arise from a combination of these two mechanisms, Hg is an evaporation residue formed principally in peripheral reactions with low excitation energies, and Po is the result of Bi activation by low energy neutrons. Fig. 8 shows that the quantities of Xe, I, He and H produced in the LBE vary only little when calculated with different models. Concerning hydrogen, this is due to the fact that it is composed principally by ^1H and therefore the differences in tritium production (and also deuterium) discussed in section 3.1 does not appear but must not be forgotten. Kr predictions differ by at most a factor 3. Deviations reaching nearly one order of magnitude are found for Ar and Ne between Bertini-Dresner and the other models.

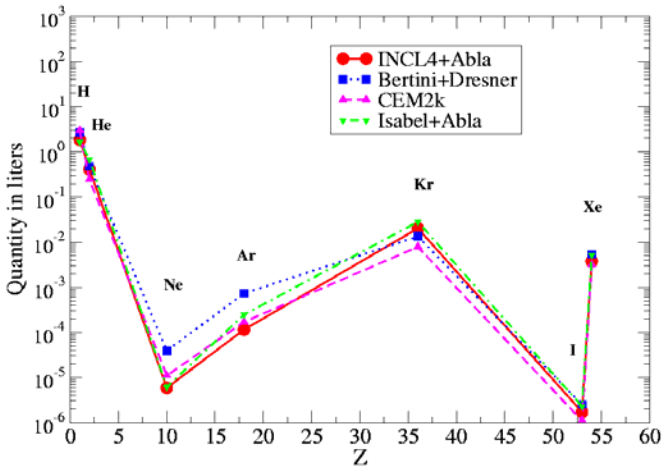

Figure 8: Quantity of gaseous elements produced in LBE using several models. These calculations were done after 123 days of irradiation with 575 MeV protons at 0.947 mA.

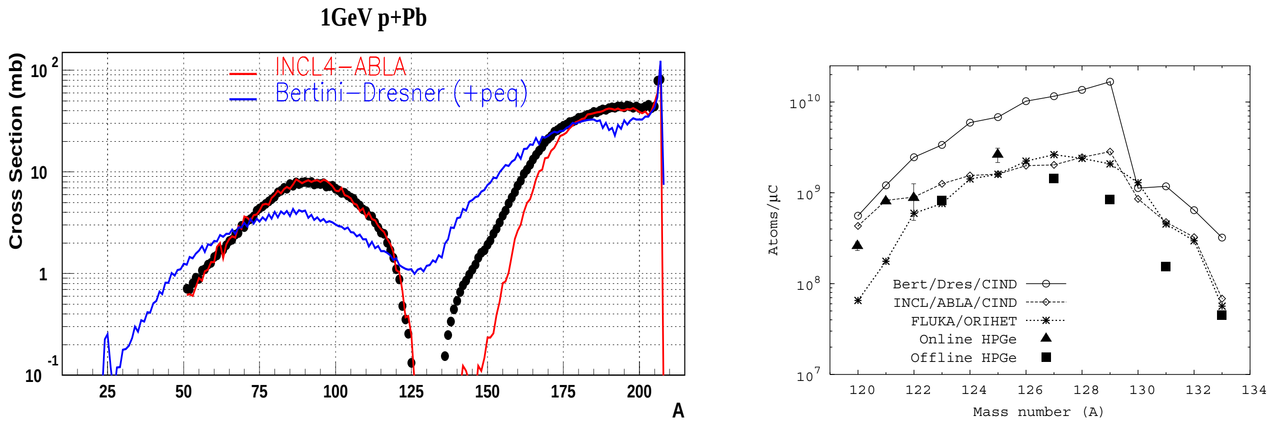

Figure 9: Left: Comparison of INCL4-ABLA and Bertini-Dresner predictions with the production cross-sections as a function of the residue masses measured at GSI for the p+Pb system [16]. Right: Production rates of Xe isotopes from a thick Pb-Bi target, bombarded with 1.4 GeV protons, measured at ISOLDE, compared to different calculation. From [18].

Comparisons of models with experimental fission yields have shown in previous works [1], that Bertini-Dresner was not predicting correctly fission fragment yields contrary to INCL4-ABLA, which is generally in good agreement with isotopic cross-sections at 1 GeV (see Fig. 9 left panel), shape of mass and charge distribution at 500 MeV and excitation functions. This has been corroborated by thick LBE target experiments at ISOLDE (CERN) [18] as shown in Fig. 9 right panel for Xe isotopes. This figure, which indicates a large over-prediction of Xe by Bertini-Dresner, could seem in contradiction with what is found in Fig. 8 where all models give similar production rates. Actually, the ISOLDE experiment was performed at 1400 MeV while MEGAPIE was irradiated by 575 MeV protons. Because spallation models may have a different behaviour as a function of incident energy, results obtained at one energy cannot always be extrapolated to a rather different energy range. This is illustrated by Fig. 10 in which a LBE target similar to the one used at

ISOLDE has been simulated with Bertini-Dresner and INCL4-ABLA at 1400 and 575 MeV. It can be observed that at 575 MeV the difference between the two models for Xe is much smaller than at 1400 MeV, explaining the results of Fig.8.

It can be also seen in Fig. 10 that fission product mass distributions are always wider with Bertini-Dresner and therefore why Ar and Ne production rates are predicted to be larger. In this case, however, it should be noted that experiments [19] have shown that Ne and Ar are generally largely underestimated by all models. This is likely because these intermediate mass fragments are only produced marginally by fission and are rather emitted in a generalized evaporation process not taken into account by the models used in MCNPX. This means that probably their production rates are largely underestimated by the models.

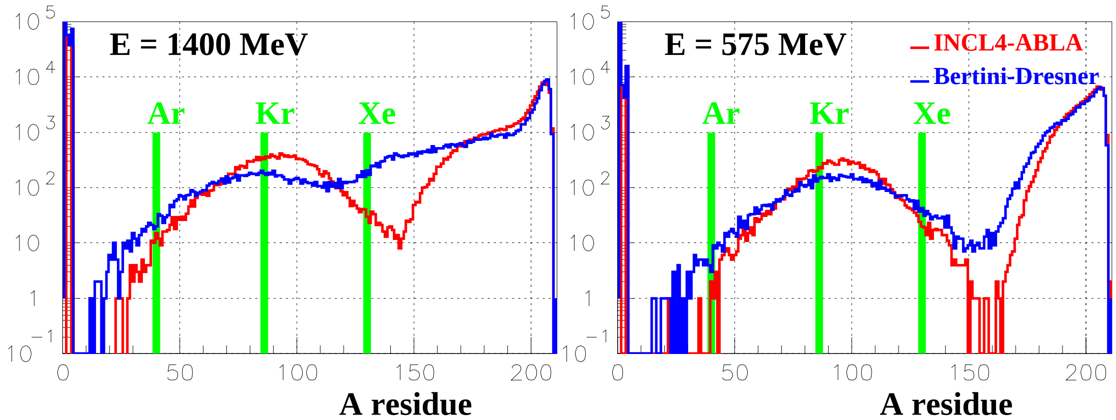

Figure 10: Production rates for Xe isotopes for 1.4 GeV protons at 600 °C target temperature. Measured points (black filled triangles and squares) compared with calculations: open circles: MCNPX (Bertini/Dresner model combination); open diamonds: MCNPX (INCL4/ABLA); stars: FLUKA. From [].

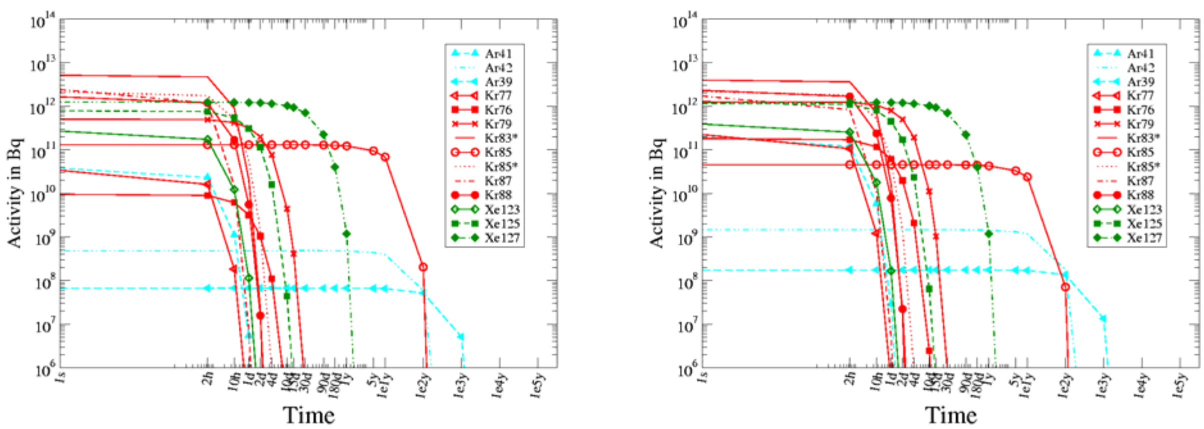

Figure 11: Main contributors to the activity in LBE coming from gaseous elements as a function of cooling time using INCL4-ABLA (left) and Bertini-Dresner (right).

Fig. 11 shows the different isotopes contributing to the activity of the gaseous elements as a function of time after the end of irradiation calculated with two different models. It can be seen that the two models give the same repartition on the different isotopes, the differences in activity level reflecting the differences already observed in Fig. 8. The activity is often dominated by Kr isotopes. Some of them (^{83}Kr or ^{85}Kr for instance) have isomeric states. While CINDER takes into account correctly the branching ratios to the different states in the decay chains, it should be stressed that the spallation models are, up to now, not able to differentiate the different states. In the case of isomers decaying preferentially by β-decay, as ^{85}Kr for example, this could change the respective rate.

During the MEGAPIE irradiation, samples of gas were taken from the LBE expansion volume and analysed by gamma spectrometry. Isotopes of Ar, Kr and Xe were found, as well as of Au and Hg. However, because the released fractions of the different gases produced are difficult to evaluate, quantitative comparisons with model calculations are impossible. Only comparisons with relative isotope production rates for a given element are meaningful.

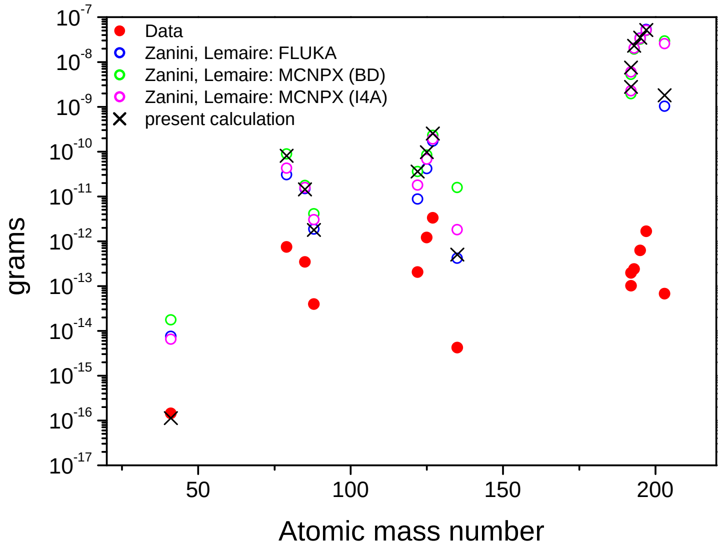

Figure 12: Comparison of different calculations with measurements of volatile elements released in the expansion volume of the LBE among which INCL4-ABLA and Bertini-Dresner denoted respectively MCNPX (I4A) and MCNPX (BD) in the legend.

3.4. Importance of impurities in the target materials

Materials always contain impurities. Even if their rates are generally low, it is interesting to study their influence on the results. Fig. 13 shows the charge distribution for the LBE and the mass distributions for the container and window of the nuclei present at the end of irradiation. The yellow points give the initial composition. In the LBE, spallation products cover nearly all possible elements. However, after the 123 days of irradiation of the MEGAPIE target their production rates are largely below the rates of the different impurities of the LBE, except for elements close to Pb and Bi. This probably means that for liquid metal corrosion problems, which can be enhanced by certain types of impurities, the spallation products are not important. However, different conclusions could be drawn for a longer irradiation time with a more intense beam.

The initial composition of the T91 container, which sees very few spallation reactions, is nearly not modified. This is different for the window in which elements with A≈35-45 and A≈85 are substantially produced.

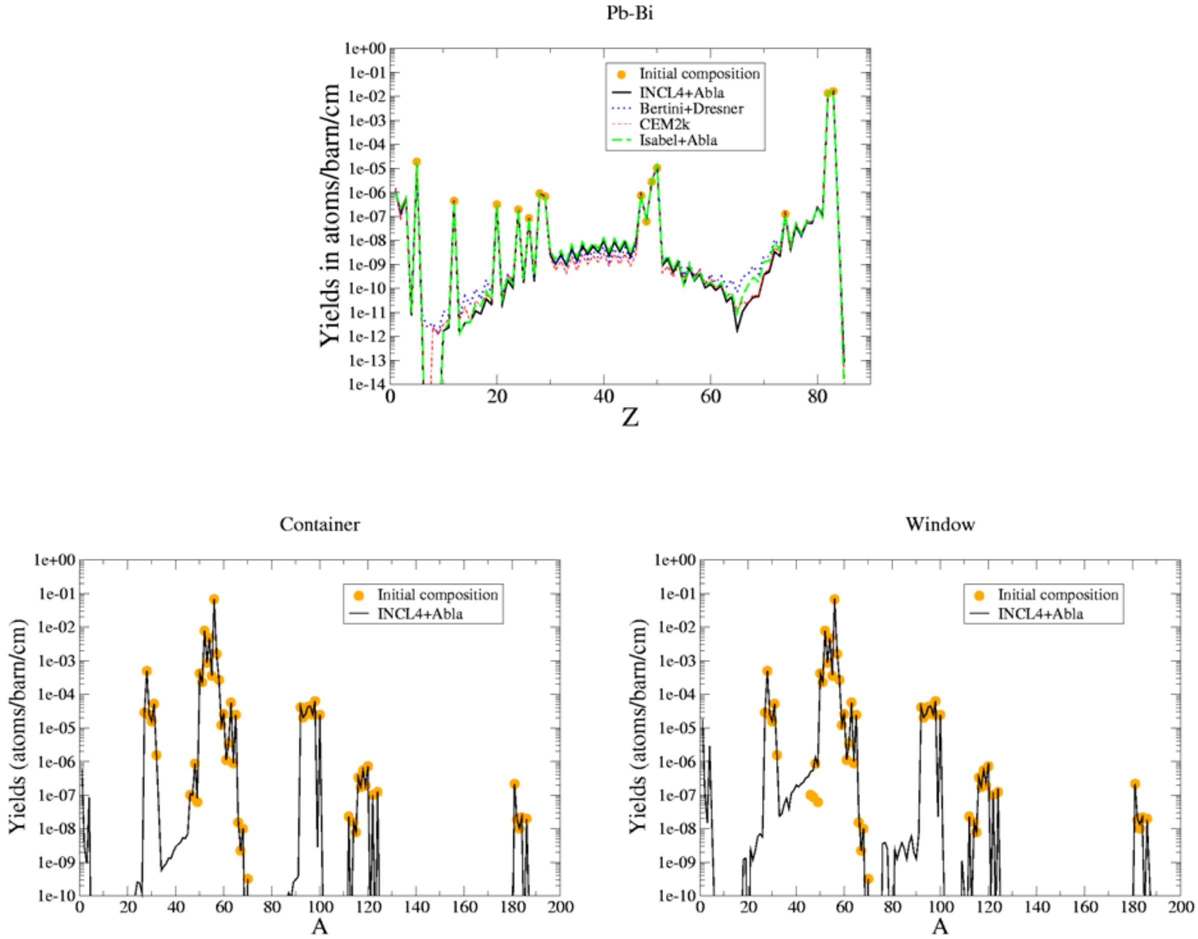

Figure 13: Concentration of elements after 123 days of irradiation by 575 MeV protons at 0.947 mA Charge distribution (top) for the LBE with the four models and mass distribution for container and window (respectively bottom left and right) with INCL4-ABLA only. Initial compositions are represented as full orange dots.

We plot on Fig. 14 activities of the different isotopes in the LBE, with and without the impurities, as a function of their atomic mass number, at different times after the end of irradiation. The calculation is done with INCL4-ABLA. It is clear here that, as far as the main contributors to the activity are concerned, the effect of having impurities in the LBE is not very important whatever the time after the end of irradiation. The only radio-isotope coming from impurities with a contribution to activity around 5% is ^{116}In in an isomer state, due to neutron capture on ^{115}In. This isotope has a half-life smaller than 1h.

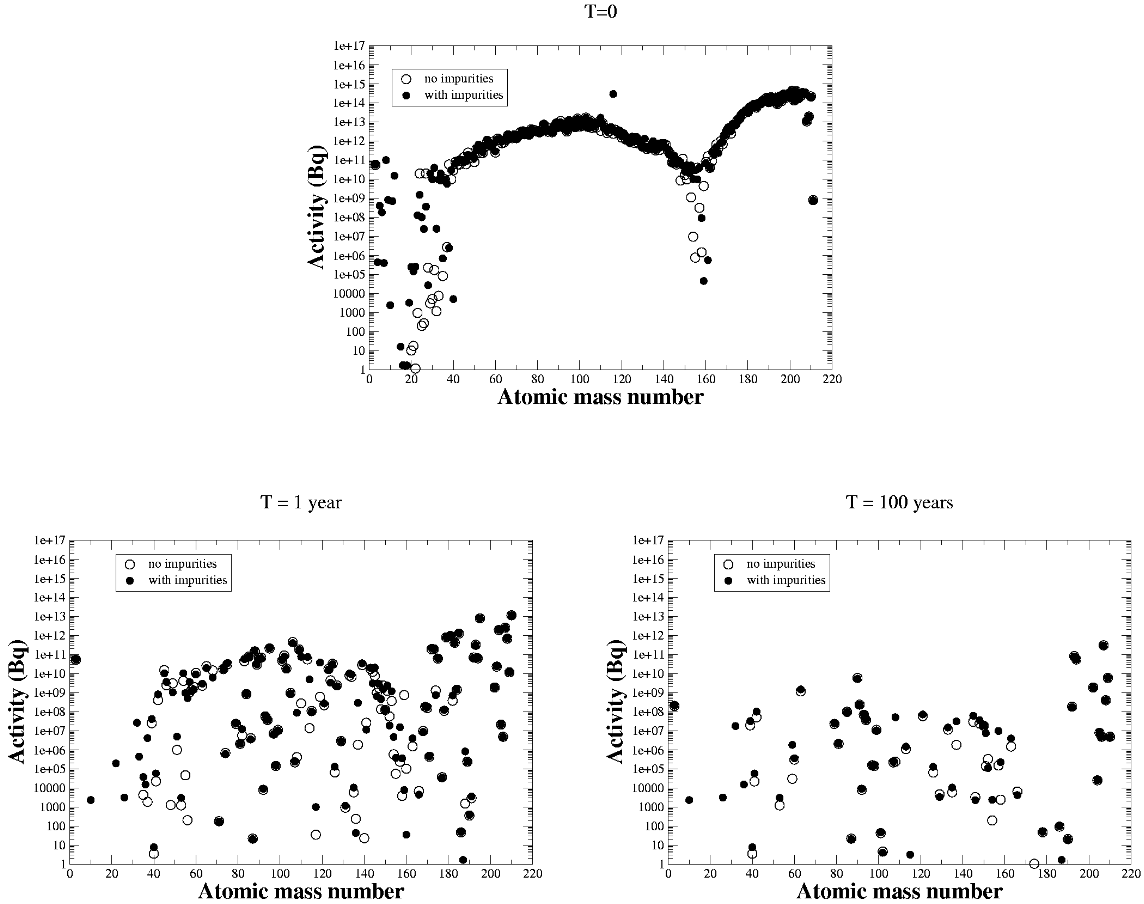

Figure 14: The activity of the different radionuclides, with (empty circles) and without (filled circles) impurities, as a function of their mass number, contained in the LBE after 123 days of irradiation with 575 MeV protons at 0.947 mA: just at the end of irradiation (top), after 1 year (bottom left) and 100 years (bottom right) of cooling time. The calculations are done with INCL4-ABLA.

4. Conclusion

Full calculations of the MEGAPIE target have been preformed in order to estimate the consequences of using the intra-nuclear cascade model INCL4 combined to the evaporation-fission model ABLA instead of the default Bertini-Dresner combination for the simulation of spallation targets. Production yields and individual activity of the isotopes produced in high-energy reactions through the models and by activation by the low-energy neutron flux have been calculated in the circulating liquid LBE, but also in the container and its bottom part the window (part between the target and the beam). Other spallation models or model combinations available in MCNPX2.5.0 have also been sometimes compared.

As already found in previous works, it has been found that generally all models give the same total activity. The main contributors are principally radioisotopes close in mass and charge to the initial nuclei. Since all models used here are known to simulate correctly the yields of these residual nuclei, the predictions of total activity can be considered as reliable whatever the model used. However, evolution with time of the activity shows that tritium, for example, can be an important contributor at a certain time. Tritium production being generally poorly estimated by the models and even, in the version of ABLA used here, not predicted at all, the results should be taken with care. The new version of ABLA should correct this deficiency. In addition, the new version of INCL4 will take into account a cluster emission during the intranuclear cascade step. This process observed experimentally is generally not treated in the models implemented into MCNPX. As shown in [20], the use of the two new models gives results in rather good agreement with elementary experimental data and therefore should lead to more reliable predictions.

Volatile elements, in particular noble gases, which are continuously emitted during the operation, have also been studied. Rather small differences with the choice of model have been found for ^{1}H or pure fission residues as Kr, Xe and I. However, since INCL4-ABLA has been shown to reproduce generally better experimental data regarding fission fragments, its use is recommended. Ne and Ar predictions vary more with the use of different models. In fact, their production yields measured in elementary experiments are generally largely underestimated by all models. This is because these intermediate mass fragments are also emitted through mechanisms not taken into account in the presently used models. This should be addressed in the new version of ABLA under development.

A last issue to be improved is the sharing of production rates between different isomers. While isomers are treated properly in the libraries in CINDER'90 in the activation by the low-energy neutron flux and in the decay chains, up to now, they are not handled by any of the de-excitation models used here.

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