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Nuclide production in spallation reactions: How useful are the simulations?

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Abstract. INCL (Liège IntraNuclear Cascade model) combined with a deexcitation code has been used a lot for numerous simulations of spallation reactions during the last two decades. We go back over some of those simulations to address the capabilities of such codes and to show some improvements. The four examples of simulation are: the EURISOL project, the MEGAPIE target, the ESS facility, and the cosmogenic nuclide production. The goal is to discuss respectively designing and optimisation, predictive power and reliability, feasibility and uncertainty estimate, and the use of modeling to mitigate lack of experimental data.

1. Introduction

Nuclide production simulations in spallation reactions are done for many years with dedicated codes. They are useful and even necessary sometimes when no experimental data are available. Four aspects are addressed in this proceedings and based on four different studies. The use of simulations to optimize the design of a facility is illustrated by the EURISOL-DS project carried out between 2005 and 2009. The reliability of the models is discussed using simulations done for the Megapie target irradiated in 2006. We tried to tackle the difficult question of uncertainty estimates within the ESS Technical Design Report (2010). Finally we show that simulations are required when no experimental data exist with the example of meteorite study.

Most of the calculations presented here are done with the intranuclear cascade model INCL combined to the deexcitation code Abla. It is also the opportunity to show some improvements done in those two codes during the last years, as well as the on-progress studies. However, the topics mentioned here are general and apply to all spallation models.

2. Optimisation: EURISOL-DS

The EUROpean Isotope Separation On-Line radioactive ion beam facility Design Study (EURISOL-DS) was a project aiming at determining the main parameters of this future european facility dedicated to the study of exotic nuclei never studied before, while taking into account the two constraints: safety and cost. The isotopes are produced in two types of target: direct (primary proton beam induced spallation reactions) or fission (the primary beam delivers, from



a converter target, a neutron flux, which induces fission in a surrounding uranium target). For our purpose we focus on the direct target.

Our goal was first to calculate the production rate of isotopes given by NuPPECC from targets using different materials and of different dimensions, with different energies for the primary proton beam, in order, in a second step, to define the best configurations. Five materials were chosen (an oxyde (Al_2O_3), a silicate (SiC), a molten salt (Pb), a refractory (Ta), and a heavy element to get heavy isotopes (UC3)). The targets were assumed to be cylinder and for each material four length and four radii were studied. The four possible energies were 0.5, 1, 1.5 and 2 GeV. Thus 320 configurations were run with two spallation models, both within MCNPX: INCL4.2-Abla ([1, 2]) and CEM2k [3] .

This method, associated to specific benchmarks of the models used, allowed to obtain the type of results displayed in Fig. 1, with the isotopes, the optimal material to produce them, the production rate given by the two codes and, not only the statistical error, but also, and surely more important, some comments on the results. Thus, for example, it was mentioned that, for the isotopes obtained after a long evaporation process, INCL4.2-Abla underestimated the rates whereas CEM2k overestimated them, or, that a specific benchmark should be done for light targets, when experimental data will be available, because results were different, with no way to disentangle between both models. Those remarks were done first to better define the reliability of the results and second to motivate future improvements in codes and/or to carry out new experiments.

Isotope	Optimal target(s) for $E = 1$ GeV	Maximal yield [atom/s]		MCNP statistical error [%]		Additional remarks
		INCL4/ABLA	CEM2k	INCL4/ABLA	CEM2k	
^9Li	Al_2O_3 ; SiC	$2.5 \cdot 10^{11}$; $1.1 \cdot 10^{11}$	$4.5 \cdot 10^{11}$; $1.3 \cdot 10^{12}$	3; 4	2; 1	Remark (1)
^{11}Li	Al_2O_3	$1.8 \cdot 10^9$	$3.6 \cdot 10^{10}$	30	6	Remark (1)
^7Be	SiC	$1.2 \cdot 10^{13}$	$1.2 \cdot 10^{13}$	< 1	< 1	Remark (1)
^{11}Be	Al_2O_3	$1.3 \cdot 10^{11}$	$4.4 \cdot 10^{11}$	3	2	Remark (1)
^{12}Be	Al_2O_3	$3 \cdot 10^{10}$	$2.3 \cdot 10^{11}$	7	3	Remark (1)
^{17}Ne	SiC	$1.3 \cdot 10^{10}$	$3.7 \cdot 10^9$	30	20	Remarks (2), (3)
^{18}Ne	SiC	$1.9 \cdot 10^{12}$	$8.6 \cdot 10^{10}$	< 1	5	Remarks (2), (3)
^{25}Ne	Al_2O_3 ; Pb	$1.2 \cdot 10^{10}$; $3.7 \cdot 10^8$	$2.4 \cdot 10^9$; $3.9 \cdot 10^9$	13; 100	25; 30	Remark (3)
^{20}Mg	SiC	$2.3 \cdot 10^{11}$	$2.4 \cdot 10^{10}$	3	8	Remark (3)

Figure 1. Part of the results obtained during the EURISOL-DS project on the isotope production rates and their optimization in material and dimension. More details in [4].

Such a study is not achievable with no spallation models implemented in transport codes and the efficiency depends strongly on the reliability and quickness of computing, two often conflicting features.

3. Reliability: Megapie

More than the reliability by itself, it is what we can do when a tool is considered reliable enough that is addressed in this part with a result obtained within the Megapie project.

Megapie was a liquid Pb-Bi target prototype irradiated for four months in 2006 at the SINQ facility (PSI). Our goal was to estimate the nuclei produced in the target and in other parts (e.g., window) to assess the safety issues related to activation and volatile release. Regarding the latter point a dedicated experiment was carried out and performed at ISOLDE (Cern) to study the release of volatiles [5].

A cylindrical (L=20 cm, r=1 cm) Pb-Bi target was irradiated by a proton beam and mass distributions of numerous volatiles (He, Ne, Ar, Br, Kr, Cd, I, Xe, Hg, Po and At) were measured. This was also used to benchmark the calculation models. All elements were rather well reproduced, except one: astatine. No model was able to simulate the right shape of the mass distribution, with several order of magnitude for the light isotopes compared to the experimental data. Since some isotopes of astatine decay in Po and astatine is more volatile than Po, this element is important for safety reasons. Thus one tried to understand and solve the simulation problem.

Astatine from bismuth can be produced in two ways: i) the light isotopes are produced by interaction of the primary proton beam on bismuth with emission of neutrons and one and only one pion, and ii) the heavy isotopes are produced by a two-step process where helium nuclei, first emitted from primary reactions, induce astatine production on bismuth. At this time the INCL4.2 cascade was unable to emit helium and the AbLa deexcitation code evaporated only alpha (no ^3He). New improved and extended versions were developed and then used, still within MCNPX. INCL4.6 [6] replaced INCL4.2 with emission of nucleons, pions and all light nuclei with mass below 8, with a nuclear potential for the pion and a special care for the low-energy reactions (case of the helium-induced reactions involved in the case of astatine). AbLa was moved to AbLa07 [7] with the possibility to emit all types of nuclei.

Those new versions were tested on the elementary processes playing a role in this study and the results were satisfactory, especially for the two-steps channel. However, the mass distribution remained badly reproduced and we had to make an assumption to be able to fit the experimental data. Since INCL4.6 combined to AbLa07 was able to give good results regarding the elementary processes, the calculation results were probably not the ones to compare to the measurements. Noting that the measured production rates and the mass, so the half-life, of the isotopes were correlated, on the one hand, and, on the other hand, the calculations were usually done in the target because the release of the most common elements were instantaneous, one decided to add a release time to the astatine, for which no information were available. Doing so and testing several times, the models could fit the data with a time between 5h and 10h, as shown in Fig. 2

When a tool is reliable enough, as it is the case for some spallation models now, it can help determining parameters beyond its own domain. This has been demonstrated here, with the possible range of values for the astatine time release in a liquid lead-bismuth target, obtained thanks to a model benchmarked and validated in the domain of interest. Reliability is a goal to reach by the model developers and a necessary step to make the tool useful for a larger community.

4. Uncertainties: ESS

A calculation result given without its uncertainty has no meaning. Determining uncertainties is much more difficult than computing a result. That is probably the two reasons why spallation models are benchmarked before to be used. This empirical way defines the validity in a given domain.

Within the ESS Technical Design Report [9] the total activities in the spallation target had to be calculated and the related uncertainties estimated. This was the opportunity to try to go beyond the usual benchmarks. Calculations were done with two models, INCL4.6-AbLa07 ([6, 7]) and CEM03 [10], implemented in MCNPX. A first, but crude, idea of the uncertainties was the comparison of both calculation results. If both models were basically identical, the uncertainties (differences) would come for coding. However, no model are identical and numerous verifications are done to strongly reduce this kind of uncertainty. The goal was then to find out the origins of uncertainties.

The activity comes from many isotopes, but fortunately only few of them are strong contributors (say higher than 1%). Thus uncertainties are supposed to be in the parameters

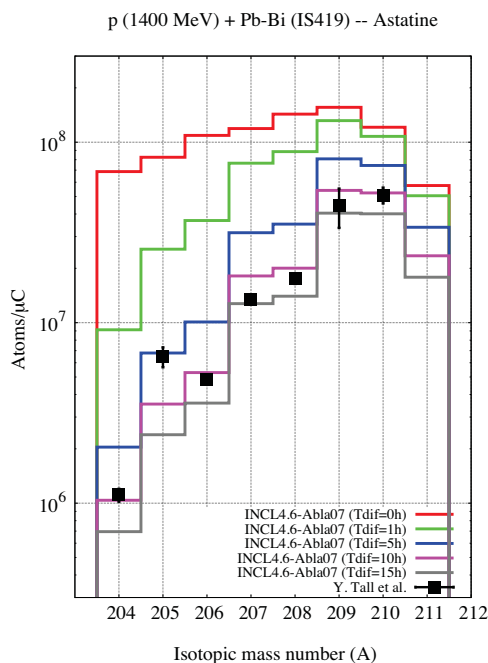


Figure 2. Mass distribution of the astatine isotopes released from a liquid Pb-Bi target irradiated by a 1.4 GeV proton beam. More details in [5] for the experiment and [8] for the modeling.

describing the contributor production. An isotope is produced by interaction of a particle with a target nucleus. Then, the particle flux and the elementary production cross section are the two parameters. Only neutron and proton play a significant role as projectile, the other particles (π , α , ...) can be neglected a priori, and the nucleon fluxes are known to be well simulated by the models. The uncertainties coming from the projectile fluxes are considered as negligible. Estimates of uncertainty of the cross sections are based on experimental data. A ratio is calculated between calculation and experiment and this ratio, depending on the projectile energy, is convolved with the particle spectrum. Several difficulties arise from this method. First, experimental data are not always available, and *similar* reactions must be sometimes used, second, data are sometimes not uniformly distributed in energy and interpolation and extrapolation are necessary, third, the data deal with proton almost exclusively. Concerning neutron, three domains of energy were defined. Below 25 MeV calculations were done with the evolution code CINDER [11] that uses a database for the cross sections, and so uncertainties were assumed to be negligible. Between 25 MeV and 200 MeV the experimental data are very scarce, which is an issue and reduces the validity of the method. Finally above 200 MeV neutron-induced and proton-induced cross sections are supposed to be the same. A schematic view of the method is given in Fig. 3

When applying carefully this complicated and tricky method, a correction of 12% was done on the target activity after nine years of cooling. All details can be read in [12] and [13]. However, having in mind the definitions taken from the GUM[14] of *uncertainty* (parameter, associated with the result of a measurement, that characterizes the dispersion of the values that could

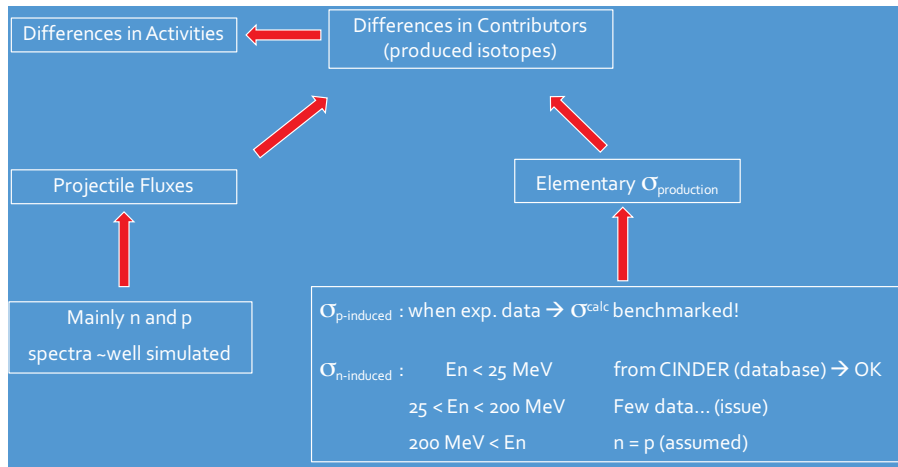


Figure 3. Summary of the quantities responsible of uncertainties in the ESS target activity.

reasonably be attributed to the measurand) and *error* (result of a measurement minus a true value of the measurand), it is clear that this method enables a correction of the calculation result based on experimental data, and so reduces the error, but does not give at all an estimate of the uncertainties. Actually uncertainty is related to precision while error is related to accuracy. Both are important, useful and difficult to estimate, but they are assessed by different ways.

5. Lack of experimental data: meteorite study

It has been said in a previous section that the spallation models, when reliable enough, are useful to study other domains than nuclear reactions (it was material science and time release). In this section that is the use of spallation models as cross section generator which is addressed.

The study of meteorites can help to get information on the primordial solar system. Meteorites are interstellar bodies, originated from asteroides, and irradiated by cosmic rays during their travel. This irradiation produces cosmogenic nuclides and astrophysicists compared those measured production rates (*PR*) to calculated ones to obtain information on the history of the meteorite and/or the cosmic ray spectra as well. The equation used below exhibits the need of elementary nuclide cross sections.

$$PR = \sum_{i=1}^N c_i \frac{N_A}{A_i} \sum_k \int_0^{\infty} \sigma_{i,k}(E) J_k dE,$$

with *i* a target element, *c_i* its concentration and *A_i* its mass number, *N_A* the Avogadro’s constant, *k* the type of particle inducing the reaction (primary and secondary particles), $\sigma_{i,k}(E)$ the production cross-section of the isotope by interaction of the particle *k* with the target *i* and with an incident energy *E*, and finally *J_k* the density flux of the particle *k*.

When available, the experimental cross sections are used, otherwise reaction models provide the missing data. Galactic cosmic rays are mainly made up protons (~87%) and helium (~12%) and their spectra are peaked in the spallation region that is roughly around 500 MeV, even if the range is very large. Then spallation models are good candidates to provide the needed cross sections. Three points can be mentioned. First, most of the experimental data deal with proton-induced reactions, but some models were significantly improved and they are now able to fit rather (sometimes very) well the data. The second point is about α , where the data are

very scarce. Astrophysicists apply then a crude approach, i.e. they assume the result of an α -induced reaction as the sum of the result of two proton-induced reactions and to neutron-induced reaction, assuming that neutron and proton give the same result. Finally, the case of neutron-induced reactions. Those reactions are secondary reactions in the meteorites where numerous spallation neutrons are produced. Here again data are very scarce and we can mention the article of Leya and Michel [15], explaining how they extract those elementary cross sections from results on a thick target, where the proton contribution is removed and with the help of a guess function. Below are given three plots (Fig. 4(2) and 5(1)), one for each case, to show how INCL4.5[6]-Abla07 compares with the experimental data and the potential use of such models to mitigate the lack of data in studies like the meteorites.

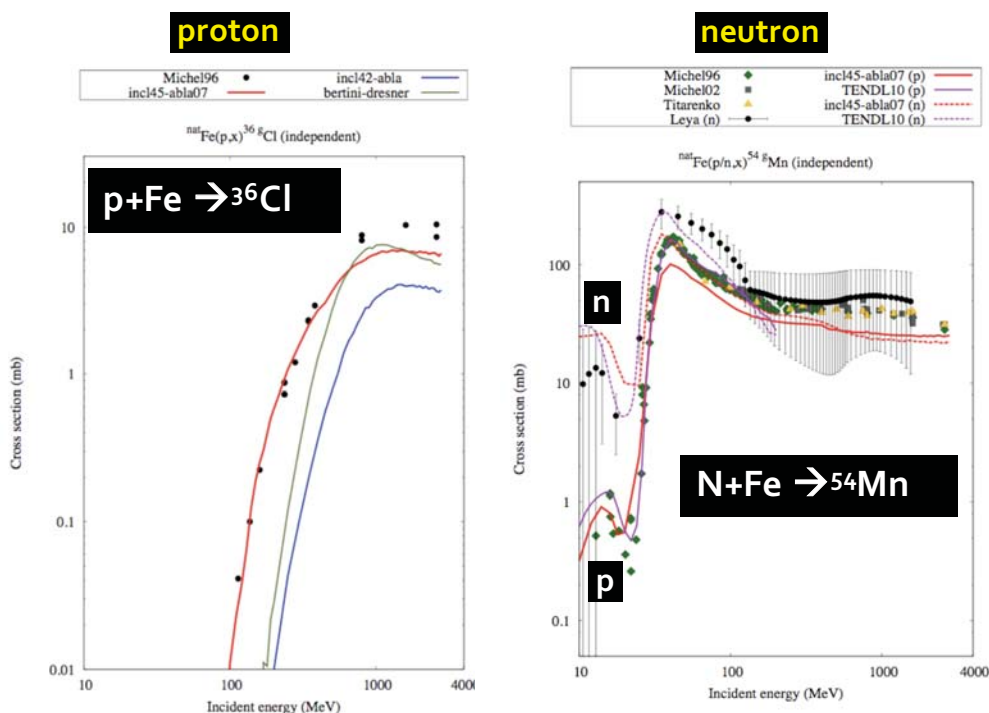


Figure 4. On the left, ${}^{36}\text{Cl}$ production cross sections from proton-induced reaction on natural Fe. More details on the figure and in [16] (page 100). On the right, ${}^{54}\text{Mn}$ production cross sections from neutron- and proton-induced reactions on natural Fe. More details on the figure and in [17].

6. Conclusion and Outlook

Nuclide production simulation in spallation reactions can be done by numerous models in several transport codes (for example see Table 1 in [13]). For the last twenty years significant improvements were done in this domain and some models can be considered particularly reliable. However, reliability, uncertainty, error are still difficult questions to answer clearly. This is the reason why benchmarks are even now needed. Some are done by the transport code developers (MCNPX, GEANT4, PHITS), some others performed in dedicated project ([18]), and sometimes must be done specifically.

Nevertheless, models are still improved and extended. For example, in the intranuclear cascade INCL, a specific study of few-nucleons removal has been initiated by D. Mancusi

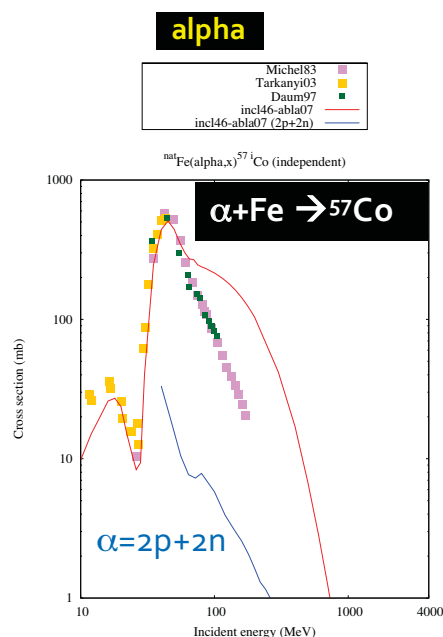


Figure 5. ^{57}Co production cross sections from α -induced reaction on natural Fe. The blue line is obtained by multiplying by four the result with a proton projectile (see text).

in [19] and is being generalized. Another extension is the implementation of strangeness (kaons, hyperons) to better describe the high-energy domain and extend the capabilities of INCL to kaon and hypernucleus production. These works are expected to be included in the next version of Geant4. The first results, compared to experimental data and other models, are encouraging.

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