

**OPTIMISATION OF A CODE TO IMPROVE SPALLATION YIELD
PREDICTIONS IN AN ADS TARGET SYSTEM**

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Abstract

In an accelerator-driven system (ADS), a target system is incorporated. The main purpose of the target system is to produce excess neutrons, which brings the subcritical reactor core in an ADS to critical condition. Critical condition allows for operation as a reactor system specifically to fulfil transmutation capability as well as electricity generation. From a safety point of view, the target system is quite a new component and potentially provides a novel radiotoxic burden to the environment and to humans. It is generally understood that there is still unknown physics involved in the operation of a target system, in particular, regarding nuclear spallation. Moreover, we have not yet developed the related engineering to cope with so-called “spallation products.” As a primary step towards better comprehension, this paper describes how we can have a better computational tool to predict the yields of spallation products in a proposed target system.

Introduction

Spallation neutron source is made with an intense beam of high-energy particles hitting a thick target made of heavy nuclei in an accelerator-driven nuclear transmutation system. As a by-product of this process, a lot of spallation isotopes are produced. The produced spallation isotopes show extensive distribution regarding mass number and lifetime. It is understood that there are some spallation products that have radioactivity comparable with Po, which is considered hazardous to humans and is followed attentively in the case of a Pb-Bi cooling system. In this regard, the amount of spallation yield must be evaluated with accuracy.

We developed a code to search a set of optimum parameters that is incorporated in the evaluation code to be compared with experiments. This optimisation code was combined with the evaluation code in order to increase accuracy of prediction of spallation yields.

We will show that the optimisation system combined with the particle and heavy ion transport code system (PHITS) [evaluation code] improved the accuracy of prediction of spallation product yields.

Computational simulation of spallation experiments by PHITS code

We used NMTC series code for the transportation of high-energy particles (including the spallation process) and we introduced the ATRAS code system. In ATRAS, NMTC/JAERI97 [1] was incorporated: the evaluations of total cross-sections below 100 MeV and elastic cross-sections are improved, ISOBAR model including the pre-equilibrium process for the intra-nuclear cascade model is introduced, and the function of level density is improved. Then, the jet AA microscopic transportation model, by which we calculate the hadron cascade process, was introduced into NMTC/JAERI97 (called NMTC/JAM). One of the crucial improvements for NMTC/JAM was the incorporation of the general evaporation model (GEM) [2] that can simulate the evaporation and fission processes in order to model particle injection from excited target nuclei. This has been acknowledged to be quite effective for the evaluation of spallation with much improved precision. For NMTC/JAM, the following improvements were incorporated into the PHITS code: the transportation of heavy ions, the quantum molecular dynamic model and the transportation of low-energy particles.

The recent development of the PHITS code has been focused mainly on heavy ion transportation and the effects of magnetic fields and gravity. Thus, the comparison between code predictions and experimental measurements was not performed for the intra-nuclear cascade ~ 1 GeV, particle evaporation in the pre-equilibrium process and from the equilibrium excited state, and the fission model, which are our major interests for the evaluation of spallation products in the ADS target system.

Our primary objective of the current study was to optimise the parameters prepared in PHITS and to reproduce the experimental data that were recently generated by GSI [3].

To measure the discrepancy between calculated predictions and experimental data, we employed the so-called “F-value”, which is defined below:

$$F = 10^{\sqrt{\langle f \rangle}} \quad \text{and} \quad f = \log(\mathcal{S}_{cal,i} / \mathcal{S}_{exp,i})^2$$

where \mathcal{S}_i is the production cross-section of the i^{th} nuclide produced by spallations in the target. The suffixes “exp” and “cal” mean experimental measurement and calculated value, respectively. In the PHITS code, there are variations in applied intra-nuclear cascade models, evaporation models and the energy level for the intra-nuclear cascade model as well as the intermittent use of the Coulomb

diffusion model. Thus, there were numerous sets for the associated input parameters. We needed to choose a technique to find the optimum sets of input parameters in order to obtain the overall most preferable production cross-sections. For this purpose, we used the method of *maximal likelihood function*. The maximal likelihood function is defined as follows:

$$L(\overset{fi}{q}, \overset{fi}{s}_{phits}, \overset{fi}{d}) = \frac{1}{(2p\overset{fi}{d}^2)^{n/2}} \exp\left[-\frac{(\overset{fi}{s}_{ex} - \overset{fi}{s}_{phits}(\overset{fi}{q}))^2}{2\overset{fi}{d}^2}\right]$$

where $\overset{fi}{d}$ is the experimental error matrix, which will be a diagonal matrix when there is no correlation among errors, and $\overset{fi}{q}$ is a vector of input parameters. This likelihood function signifies the probability that the calculated production cross-sections agree with the experimental measurements. In order to determine the maximum of the function, we use the logarithmic maximal likelihood function defined by:

$$M(\overset{fi}{q}) = \log L(\overset{fi}{q}, \overset{fi}{s}_{phits}, \overset{fi}{d}).$$

The adopted parameters to optimise are: the parameter for the calculation model choice of 14, which takes the discrete values, and the five continuous parameters. Degree of freedom was 256 for the 276 experimental data that we used.

Global optimisation method

We used the maximal likelihood method to optimise these parameters, utilising the likelihood function made of the production cross-section for each isotope produced by spallation. In order to search global maxima for the likelihood function, the DIRECT algorithm [4] was used, which is known to be a very efficient method for optimisation. In this algorithm, we can find the maxima with better accuracy by automatically dividing the parameter space to trap the point of the maximal value. Figure 1 shows the schematic image of narrowing down the space to the subspace where we can find the maximal point. Among the provided experimental data, we used selected data of isotopes produced with high production cross-sections. This data was chosen because the calculated statistical error grows for isotopes produced with small production cross-sections. For each element, we centred the isotope with the largest yield. Then, for comparison with the experiments, we weighed the isotopes by a Gaussian distribution having the width of neutron numbers with statistical accuracies of greater than 0.1 under the same conditions. By this method, we performed the optimisation for all nuclides with mass numbers from 40 to 200. There are two ways of optimisation – one is global optimisation over all elements and the other is local optimisation for a specific element.

Figure 2 shows the results obtained for ${}_{60}\text{Nd}$, which is one of the rare earth elements of interest to us. This case is optimised specifically to Nd. As the figure indicates, the experimental measurements were very well-reproduced. However, the overall χ^2 value was 659, which means it is a less preferable reproduction of the experimental measurements as a whole.

Figure 1. Schematic image of dividing the parameter space through the DIRECT algorithm

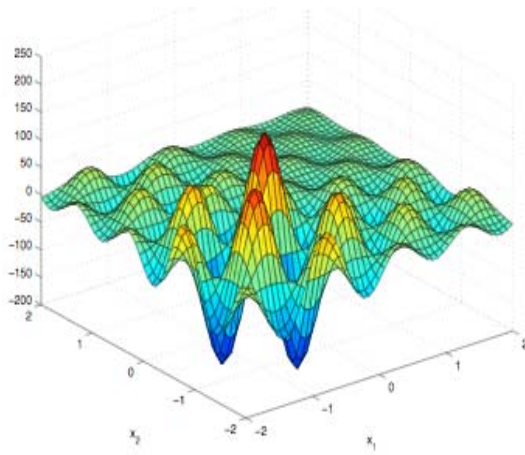


Figure 2. Production cross-section of ${}_{60}\text{Nd}$ in the reaction ${}^{208}\text{Pb} + p$ (1 GeV) obtained through experiments and simulations

(a) CASCADE code simulation, (b) PHITS code simulation without optimisation and (c) PHITS code simulation with optimisation

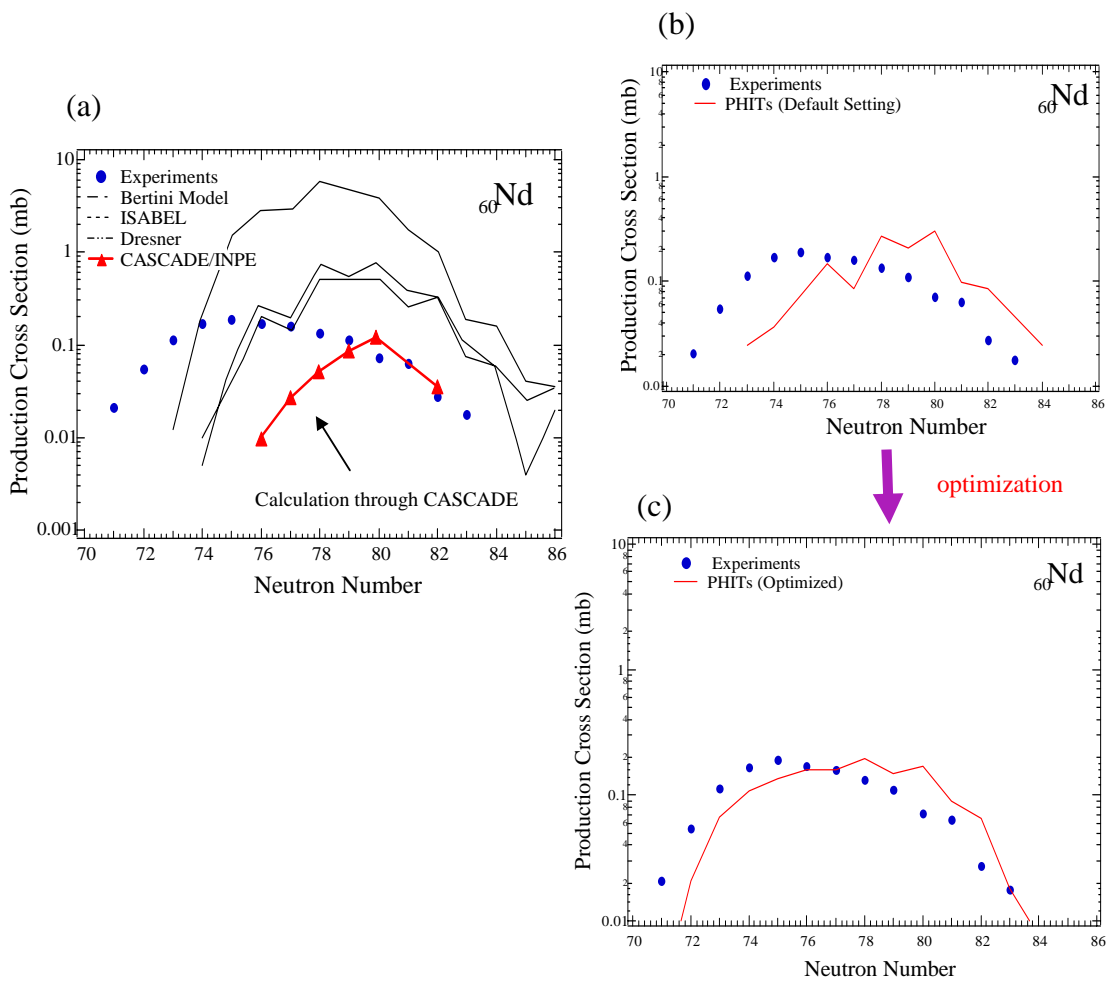


Figure 3 shows the results of global optimisation with the χ^2 value of 397. As shown, the reproduction capability for nuclides with atomic numbers from 30 to 50 was very well-improved. However, when the atomic number was greater than 60, the production cross-sections for isotopes with excess protons were badly underestimated. It can be understood that injection of neutrons during the evaporation process seems to be underestimated in the experiment because it occurs in evaporation under an equilibrium state. For this study, intra-nuclear cascade calculations using the molecular dynamics model were not performed. This was due to the very long CPU time required and the division of parameter space not being precise enough to produce satisfactory results. In any case, we think that our work has shown that the neutron emission model during evaporation should be improved.

**Figure 3. Production cross-section in the reaction $^{208}\text{Pb} + \text{p}$ (1 GeV).
Results of the PHITS code simulation with global optimisation.**

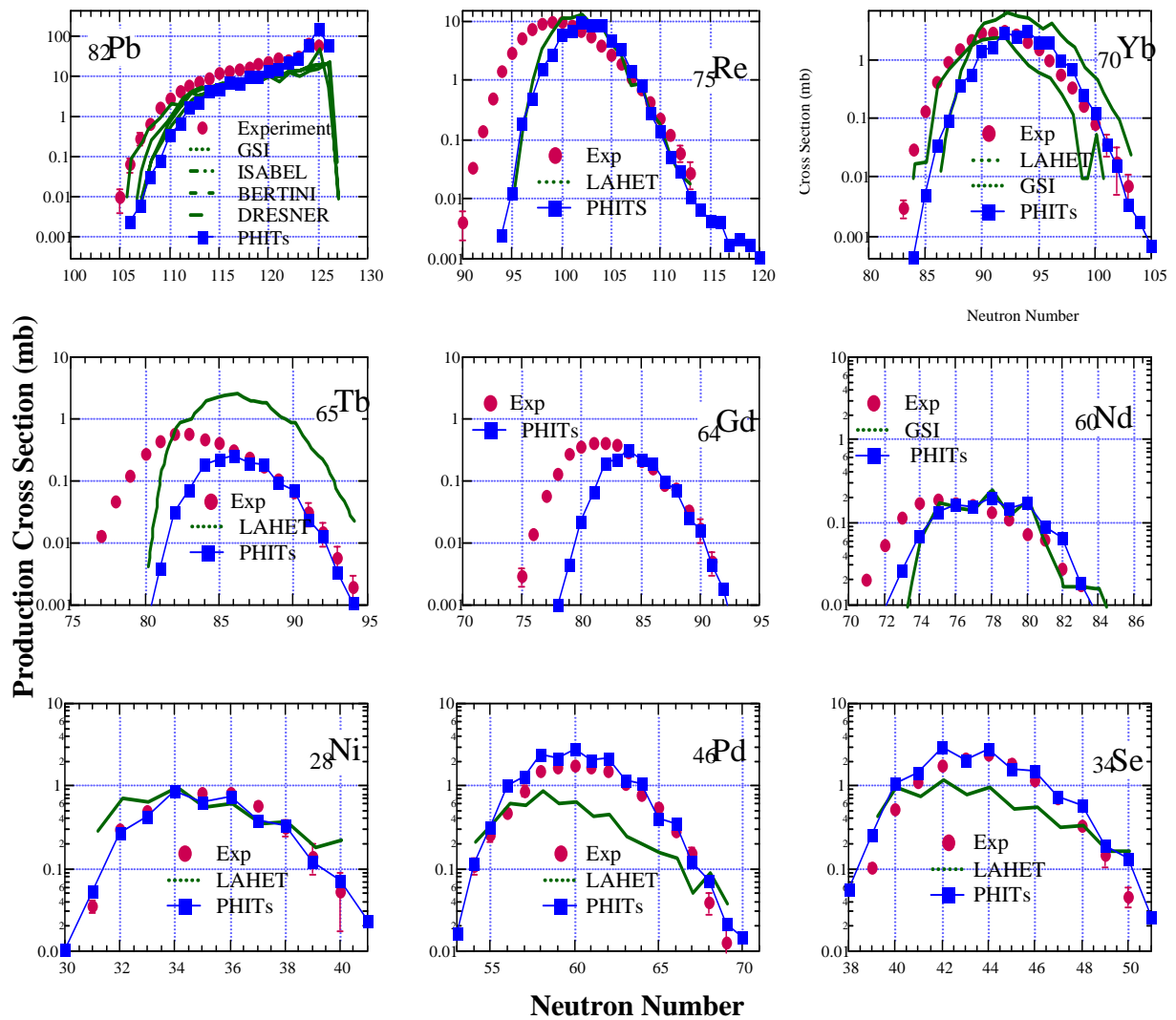
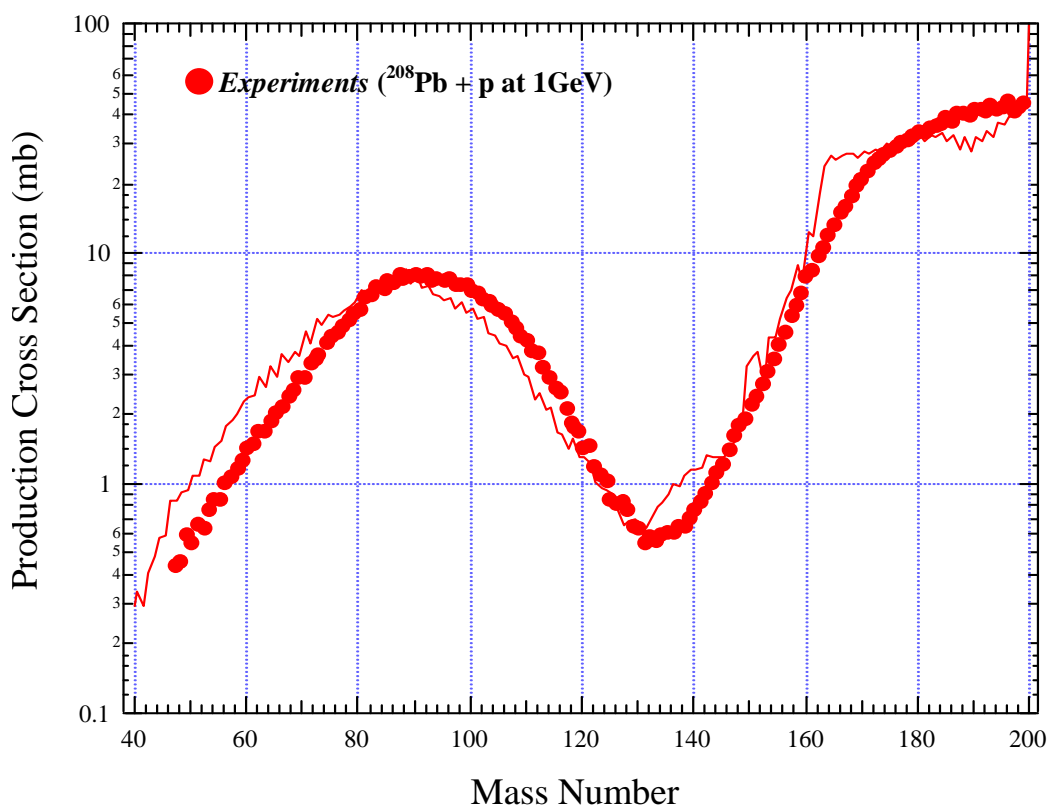


Figure 4 shows the comparison between calculations and experiments over all mass numbers. For this figure, we integrated the production cross-sections over all the produced isotopes with the same mass number. Here we found some problematic discrepancies for the rare earth region and elements with mass numbers less than 80.

Figure 4. Production cross-sections integrated over all produced isotopes with the same mass number and obtained through a PHITS code simulation with global optimisation (red line) and experiments (filled red circles)



Discussion

As seen in Figure 3, for elements with an atomic number larger than 60, the calculated results were rather shifted to the isotopes of larger mass numbers, so that the production cross-sections of excess proton isotopes were underestimated. This simply means that the neutron emission from residual nuclei was smaller compared with the experiments. If we consider that almost all neutrons are emitted during the evaporation process, we can guess the following reasons for the discrepancy.

The simplest reason may be that the excited energy for the transition to the evaporation process is too low, such that the condition of the emission of neutrons during evaporation is not well-developed. In order to resolve this issue, we need to incorporate the pre-equilibrium process and leave enough excited energy to promote neutron emission during evaporation. Raising the excited energy at the final stage of the cascade process would also be effective. This can be done by setting a smaller value for the energy cut-off parameter in the cascade process. In fact, we confirmed that this is effective in increasing the production of proton-excess isotopes. Unfortunately, energy cut-off was not an input parameter for PHITS code but a built-in one, and thus not included in the global optimisation procedure.

The other possibility is that the discrepancy may be improved by considering neutron emission in the transition to fission, with the formation of multiple fragments of residual nuclei in the range of excited energy ~ 4 to 5 MeV. It should be noted that we must re-evaluate physical parameters used in the evaporation model if we modify the models preceding the evaporation process. The fact that in Figure 4 we observe a discrepancy between experiments and calculations for nuclides where the mass number was less than 80 indicates the drawback in the evaluation of the fission process.

Figure 1 shows the comparison of experiments and predicted production cross-sections of PHITS with optimum parameters.

Summary

Via the above-described optimisation method, we obtained better prediction capability using the PHITS code for overall yields of spallation products generated and accumulated in an ADS target system. In particular, the predicted cross-sections for the low mass isotopes were in good agreement with experiment data. On the other hand, the discrepancy for high mass isotopes was relatively large. From this study, we found that the predicted cross-sections were sensitive to the parameters that determine the energy at which the adopted intra-nuclear cascade model was changed or at which the evaporation model was to be adopted. For even better prediction capability, we need to further improve excited energy for the evaporation process, which is currently underestimated, and the model for the fission process.

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TECHNICAL SESSION III

Subcritical System Design and ADS Simulations

Chairs: W. Gudowski, H. Oigawa

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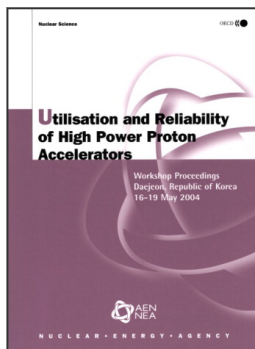
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