Combining Semi-Classical and Quantum Mechanical Methodologies for Nuclear Cross-Section Calculations Between 1 MeV and 5 GeV

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With a goal to develop a nuclear cross-section code usable over the wide energy range of 1 MeV to 5 GeV, one option is to combine intranuclear cascade, pre-equilibrium, and Hauser-Feshbach models in existing codes. However, the first two models are semi-classical while the third one is quantum mechanical, and combining them is not straightforward because the third model requires spin and parity distributions for all excited states that cannot be supplied by either one of the first two models. Approximations to overcome this difficulty are described in this paper. Success of this combined model will allow nuclear data evaluations for a large number of materials whose cross sections are needed in a wide range of applications, including the design, operation, and future upgrades of the SNS (1 GeV proton). The incident particles may be neutrons, protons, charged pions, or photons. Though only partially completed at this time, the new model compares well with experimental radionuclide production cross sections from thresholds to 2.6 GeV for proton-induced reactions on Fe.

KEYWORDS: nuclear model, cross sections, calculations, iron, radioisotopes

I. Introduction

Two existing model codes and additional developments described in this paper are utilized to achieve the stated goal. TNG[1)] is a low-energy (1 MeV to 40 MeV) nuclear cross-section model code developed as a tool for generating the evaluated ENDF/B neutron files. TNG is based on a unified Hauser-Feshbach (H-F) and pre-equilibrium (P-E) formalism emphasizing the importance of discrete level structure. CEM[2)] is a high-energy (40 MeV to 5 GeV) nuclear cross-section model code that uses an intranuclear cascade (INC) model, a P-E model and an evaporation model. CEM is semi-classical, hence it does not utilize discrete levels, their spins and parities, and their constraint on level density parameters. No matter how high the incident particle energy is, all residual nuclides have part of their excitations in the low-MeV range during the decay process, requiring partial wave analysis in the H-F model for further particle emission and creation of final radionuclides. The major challenge in developing a combined model code is reconciling the semi-classical physics in CEM with the quantum mechanical physics in TNG. An approximate method to solve this problem is described in this paper. Thus, a cross section calculation starting with the INC model for projectiles in the few-GeV range may pass through the P-E model in the high-MeV range, the H-F model in the low-MeV range, and finally with the gamma-ray cascade model in TNG, reach the ground states of several hundred residual nuclides. Also, the transition from a pure TNG calculation at an incident energy of 40 MeV to a combined CEM-TNG calculation above 40 MeV must be natural and smooth. Cross sections for discrete levels and for gamma-ray production will be obtained for incident energies between 40 MeV and 5 GeV.

Stated simply, the evaporation model and the low-energy end of the P-E model in CEM will be replaced by the unified P-E/H-F model in TNG. This simple statement is hard to implement because it involves a transition from semi-classical physics to quantum mechanical physics. The required input for TNG, such as spin and parity distributions for all excitation energies in all residual nuclides, are simply not available from the INC model and the high-energy P-E model in CEM. This information is required for complete and realistic cross-section evaluations for reactions induced by high energy particles.

ENDF/B-VI, the most recent version of evaluated neutron cross-section files in the U. S., allows discrete-level information to be entered for precise descriptions of the cross sections exciting each discrete level, as well as associated gamma-ray branching ratios for gamma-ray production calculations in processing codes. This type of information can only be generated by analyzing experimental data with P-E/H-F codes such as TNG. And this type of discrete-level information does exist for the 26 isotopic evaluations contributed by ORNL to ENDF/B-VI, all done with the aid of TNG analyses. This detailed information is needed by data users and will be the type of information available from successful completion of the present model up to incident energies of 5 GeV.

Since a large number of excited nuclides will be de-excited by TNG, a code automating its input for each residual nuclide, including discrete levels, their spins and parities and gamma-ray branching ratios, and reaction Q-values, has been developed.[3)] Some other elements important for combining the two codes are described. Calculated results using TNG and CEM are compared with experimental data for Fe up to 300 MeV in order to understand the successes and failures of the two codes. Emphasis is being placed at energies around 150 MeV, the upper energy of the LA150 library,[4)] to demonstrate the need for extending the LA150 library to

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higher energy. One of the reasons for limiting the upper energy to 150 MeV is that it is the pion threshold, above which more difficulties in calculations and evaluations arise. Reference 3 and the present paper complement each other.

Lillie and Gallmeier[5)] have developed a coupled neutron and photon library, HIL02k, for neutron energies up to 2 GeV. Of particular interest to the present work is that there exist discontinuities at 150 MeV in many cross sections between this library and LA150. The present model promises a smooth transition across 150 MeV for all partial reaction cross sections.

Chadwick *et al.*,[4)] in developing the LA150 library, used a global set of optical model parameters for both neutrons and protons between 30-50 MeV and 150 MeV. Koning *et al.*[6)] evaluated cross section data for the Fe and Ni isotopes with an upper energy of 200 MeV. They developed valuable optical model parameters for these elements for both neutrons and protons covering the the energy range from 1 MeV to 200 MeV. Shen[7)] determined a set of optical model parameters simultaneously for Fe and Pb isotopes up to 300 MeV. All these developments are for applications in studying accelerator-driven systems. The set of global optical parameters of Ref. 4 has been adopted as one of the default sets in TNG and used in the present work. Those in Refs. 6 and 7 will be considered in the future.

II. Model Approximations

We have validated TNG for incident energies below 40 MeV and will assume for the present paper it is satisfactory up to this energy. Above 40 MeV TNG needs a direct reaction component that we plan to adopt from the INC model in CEM. We also need the INC model to account for pion production for incident energies above 150 MeV. Therefore, the present effort is for incident energies above 40 MeV. We present first our approximation for incident energies between 40 and 80 MeV, and then for incident energies from 80 MeV to 5 GeV.

CEM and TNG are run for the same incident energy if it is between 40 MeV and 80 MeV. From CEM we write into a file the excitation spectra (cross sections as a function of excitation energy) below an excitation energy of 40 MeV in all residual nuclides. These nuclides are no longer followed in CEM. TNG reads this CEM file and continues the decay process. Because CEM is Monte Carlo, the CEM file is converted into TNG group structure first. Then the excitation spectra calculated in TNG after the first particle emission are combined with those from CEM (see paragraph below for the combination method), while replacing the part of TNG spectra having excitation energies above 40 MeV to small values. These small values are arbitrarily fixed as 0.001 mb per group (group width of 1-2 MeV) because this part of TNG is already accounted for in CEM. Now we have spin and parity distributions calculated by TNG and can proceed with the H-F model to calculate the emission of the next particle.

From the second particle on, TNG and CEM excitation spectra are summed (not combined, see next paragraph). Our first approximation is that the CEM output have the same spin and parity distributions as calculated by TNG in the excitation energy range below 40 MeV. This is an approximation because the INC model in CEM has a direct reaction component whose spin and parity distributions may be different than those calculated in TNG. The combined code is referred to hereafter as CETNG (Cascade Exciton TNG), used for incident energies above 40 MeV. Thus, from here on, excitation spectra from CEM, TNG, and CETNG have completely different definitions.

The CEM and TNG excitation spectra in the residual nuclide after the first particle emission (also called the binary reaction) are calculations from two different models for the same quantities. We combine them in CETNG by using a weight $(40/E)^2$ for TNG and $1-(40/E)^2$ for CEM where E is the incident energy. The combined excitation spectra in CETNG deexcite and produce new excitation spectra (in a daughter nucleus) that are referred to as CETNG excitation spectra. The weights are intended to smooth all calculated results across the 40-MeV incident energy between TNG (used below 40 MeV) and CETNG (used above 40 MeV). The direct component from the INC model in CEM, that produces harder particle emission spectra than TNG, also phases in smoothly in CETNG. Another advantage in using these weights is that the first-chance alpha-particle emission is extremely low in CEM, and keeping some TNG contribution to CETNG reduces this problem. After the second particle emission, CETNG excitation spectra and the new CEM input are summed (not combined) because from this emission on, the CEM part comes from the decay of nucleii having excitation energies above 40 MeV while the CETNG part arises from those below 40 MeV, hence the two components complement each other.

The replacement (to 0.001 mb per group) mentioned above for excitation energies above 40 MeV calculated in CETNG is meant to trick CETNG to calculate spin and parity distributions for daughter nucleii from mother nucleii having excitation energies up to the incident energy (limited to 80 MeV if the incident energy is greater than 80 MeV, as explained below).

A second approximation arises because TNG methodology is not appropriate for incident energies above 80 MeV. For CEM runs with incident energies greater than 80 MeV, TNG is always run with a fixed incident energy of 80 MeV. After emitting a few particles in the CETNG calculation, the maximum excitation energy in the residual nuclide may drop (due to the Q-value loss) below 40 MeV, a maximum excitation energy not high enough to accomodate the CEM excitation spectra in that residual nuclide. To prevent this from happening, CETNG excitation spectra after each particle emission are extended to 80 MeV before applying the replacement method described above. The spin and parity distributions for the extended energy region, from about 72 MeV to 80 MeV (assuming a Q-value of 8 MeV for emitting one particle), are missing and are filled by the distributions available in the next closest excitation energy. The widened energy region, being twice as wide as the needed 40 MeV, is designed to generate spin and parity distributions for the next residual nuclide. The extension is no longer needed if the extended energy reaches the kinematic limit.

The above extrapolation of spin and parity distributions in the extended excitation energy region is our second approximation, needed only for incident energies above 80 MeV. The extension of excitation energy is limited by the reaction kinematics. For example, the extended excitation energy can be at most 5 MeV if the incident energy is 85 MeV, and the extension would have reached the kinematic limit after the second particle emission, and no more extension is needed for further particle emissions. If the incident energy is 5 GeV, the extension will continue until the total excitation cross section in every residual nuclide drops below the present code limit of 0.01 mb.

III. Preliminary Results

Comparisons with experimental radionuclide production data reveal some problems we did not identify in the beginning. We chose Fe as a test case since the first author is one of the evaluators for the Fe isotopes in ENDF/B-VI. The calculated results are preliminary because we are still refining the model and because the model parameters used are somewhat arbitrary, taken from defaults built in CEM and TNG. The CEM results for incident energies up to 150 MeV were normalized to total reaction cross sections evaluated in the LA150 library. The total reaction cross sections from CEM in this energy range are too low, by up to 14%. For incident energies above 150 MeV, the total reaction cross sections calculated by CEM were used.

Comparisons of calculated radioisotope production cross sections for natural Fe with those measured by Michel *et al.*[8] for incident protons up to 2.6 GeV are shown in **Fig. 1**. TNG is used for incident proton energies below 40 MeV, CETNG is used above 40 MeV. These measured data are helpful for the present development because they cover the energy range we are trying to model and because they include the most difficult reactions to calculate. The data shown for the Co-56, Co-55, and Mn-54 productions are from the Fe-56(p,n), (p,2n), and (p,2pn) reactions, respectively. The Mn-52 production data have a low-energy peak due to the Fe-56(p,nα) reaction, a high-energy peak due to Fe-56(p,2p3n), and a small contribution from Fe-54(p,2pn). The experimental Cr-51 production data shows three peaks, the 45-MeV peak is from the Fe-56(p,pnα) reaction, the 90-MeV peak is from Fe-56(p,3p3n). Included in the calculated Cr-51 production is a small contribution from the Fe-54(p,3pn) reaction. The measured data for Cr-51 below 30 MeV is from the Fe-54(p,pHe-3) reaction that we have not yet calculated.

There are a few disagreements between our calculated data and experimental data shown in Fig. 1 that we need to examine further. For example, the calculated Co-56 and Co55 production cross sections around 300 MeV are too high by a factor of 2. The measured Mn-52 production data show a peak near 75 MeV, ours is near 100 MeV. We also miss the threshold region for Mn-54. Even with these disagreements, our results are still very good in view of the large discrepancies and limited energy ranges seen in the 29 contributions to a model code comparison compiled by Michel and Nagel.[9]

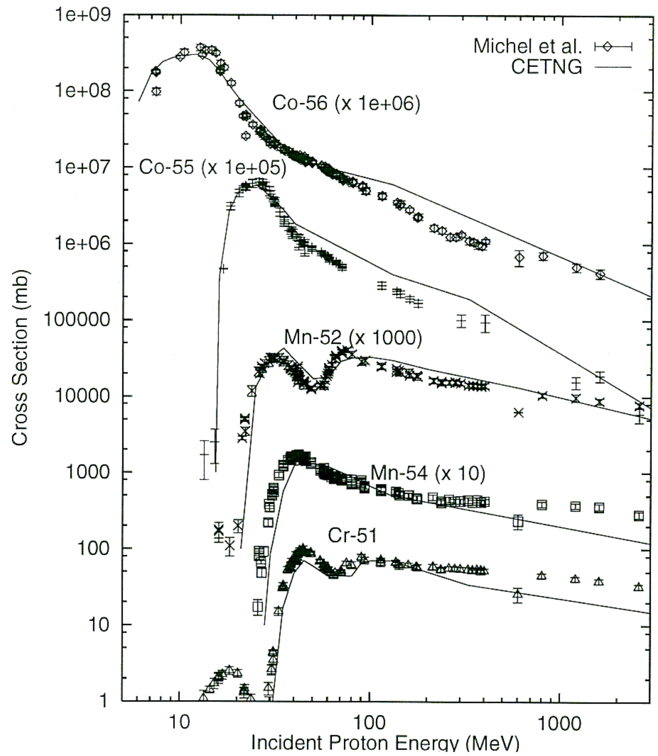

Figure 1 Cross sections of radionuclide production from proton-induced reaction on iron.

During the calculation for incident energies above a few hundred MeV, we encountered a problem. In combining the CEM excitation spectra with those of TNG, we match up t heir residual nuclide mass, charge, and reaction title such as (p,ppn). Above 150 MeV, reaction titles containing pions such as (p,pp+-n) appear and since TNG cannot recognize these titles, these reactions are skipped. The symbols + and - are used for charged pions. Skipping these reactions may cause an under-prediction of more than 10% above 1 GeV. For the calculated data shown in Fig. 1, the titles having pions have been converted to ones recognizable by TNG and to yield the correct residual mass and charge. For example, the reaction (p,pp+-n) becomes (p,ppn). A similar conversion also solved a smaller problem arising from the fact that TNG does not calculate d, t, and He-3 cross sections explicitly. Converting these particles to neutrons and protons yields correct nuclide productions but wrong particle productions. For examples, (p,d) becomes part of (p,pn), but deuteron production is missing. For correct d, t, and He-3 productions we plan to expand TNG to include them.

IV. Work in Progress

CEM has several default level-density models. We chose the model by I1jinov *et al.*[10] as it is suitable for excitation energies extending from neutron binding energies to several hundred MeV. This model accounts for odd-even effects and shell structures near the neutron binding energy and allows the shell structures to disappear above 100 MeV. TNG has a similar level-density model due to Mengoni and Nakajima[11] but TNG modifies the low energy part by taking into account the available discrete levels using the Gilbert and Cameron technique,[12] and hence is a stronger level-density model in the low energy range. We have started to unify the level-density models in the two codes by using the total number of discrete levels up to a certain excitation energy as a constraint to level-density parameter in CEM and by making the models in Refs. 10 and 11 available in both codes.

The present version of CETNG takes several steps for one single incident energy, and hence is rather tedious. We are combining these steps to run several incident energies at once, then to repeat the present calculation with more carefully chosen parameters, for example, the optical model parameters of Koning *et al*.

The present version of CETNG allows only eight decay chains, meaning we cannot produce any nuclides from the emission of more than eight particles. We are expanding CETNG to handle at least 20 decay chains.

V. Conclusions

The desired smooth transition across the 40-MeV incident energy between a TNG calculation below and a CETNG calculation above has been achieved. CETNG can yield excitation cross sections for discrete levels and associated gamma-ray production in each residual nuclide between 40 MeV and 5 GeV, not available in CEM.

There are problems we do not yet know how to solve. At an incident energy of 40 MeV, (p,p), (p,n), and (p,α) cross sections calculated with TNG and CEM differ sharply. Combining TNG and CEM above 40 MeV reduces this problem but the reduction diminishes as the incident energy rises. TNG, developed for the ENDF/B evaluations, is flexible in choosing optical-model, level-density, and P-E parameters for best fits to experimental. data, but CEM does not have such flexibities, especially the absence of α-particles in its INC model.

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