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## New High-Energy Evaluated Nuclear Data Libraries for the Accelerator Production of Tritium Program (U)

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*The Accelerator Production of Tritium (APT) program requires accurate nuclear data to model the performance of the target/blanket assembly, tritium production, activation, heating, and damage. To meet these needs, we are developing advanced nuclear-data evaluations and libraries to be used in transport calculations for APT design. Evaluations extend up to 150 MeV for important target and shielding isotopes and are being performed for both incident neutrons and incident protons. All evaluations specify production cross sections and energy-angle correlated spectra of secondary light particles as well as production cross sections and energy distributions for heavy recoils. The evaluations are based on theoretical analyses performed with the GNASH model code, benchmarked to experimental data. Evaluations will be processed into a newly-expanded format appropriate for the MCNP family of continuous-energy Monte Carlo radiation transport codes. Resulting libraries will be the first for MCNP to allow completely coupled neutral and charged-particle calculations in continuous-energy. Preliminary modifications to MCNP to support such simulations have been accomplished. (U)*

### Introduction

The Accelerator Production of Tritium (APT) program is designing a high-current proton linear accelerator to produce tritium for the United States' nuclear weapons stockpile. The design consists of a 1 - 2 GeV proton beam incident on a thick tungsten target surrounded by a lead blanket. Through (p,xn) and (n,xn) nuclear reactions, neutrons are produced and are thermalized by heavy water. These thermal neutrons are subsequently captured on helium-3, which flows throughout the target-blanket system, to produce tritium via the (n,p) reaction.

The LAHET Code System (LCS) has been instrumental in guiding the design of the APT system (Prael et al. 1989 and 1995), to predict and optimize key design parameters such as the total number of neutrons per incident proton (n/p), and the total number of tritons per incident proton (T/p). Currently the LAHET intranuclear cascade (plus evaporation / Fermi-breakup) code is used to model the nuclear interactions as well as the radiation transport above 20 MeV (and below 20 MeV for incident charged particles). Below 20 MeV, the nuclear interactions and the transport of neutral particles are handled by the MCNP code (Breisemeister, 1991) which uses ENDF/B-VI evaluated nuclear data libraries. The intranuclear cascade (INC) models in LAHET are most accurate above 150 MeV where semiclassical approximations become applicable. Below 150 MeV, however, nuclear interactions become more sensitive to specific details of nuclear structure along with quantum effects in the scattering. For this reason, we are extending the evaluated nuclear data libraries for both incident neutrons and protons up to 150 MeV using the GNASH nuclear reaction model code along with experimental data. The GNASH code

applies direct, preequilibrium, and Hauser-Feshbach evaporation theories and in a recent code comparison was shown to be one of the most accurate codes available for modeling nuclear reactions below 150 MeV (Blann et al., 1994).

To utilize these new high-energy libraries, the LCS system is being developed to include a merging of essential elements of MCNP and LAHET, along with data formats that enable the merged code system to use the new 150-MeV data libraries, allowing neutrons and protons to be transported in a fully coupled manner. The goal of this work is to improve the accuracy of LCS simulations of the APT target-blanket by using high-quality nuclear data below 150 MeV, where much of the neutron production occurs. In addition to an improved predictive capability of (n/p) and (T/p), this work will lead to more accurate predictions of shielding requirements, heating, activation, and radiation damage.

Prior to the present work, high-energy neutron and proton libraries up to 100 MeV were first developed at Los Alamos in the late 1980's for a range of elements (Young et al., 1990). More recently, high-energy libraries were generated for particle radiotherapy simulations in the Livermore PEREGRINE project, through a Livermore - Los Alamos collaboration, (Chadwick et al., 1996c). The present APT evaluated data libraries represent an advance over these earlier works since they also include spectral and yield information for the heavy recoils, as well as being based on the latest version of the GNASH code (Young et al., 1996).

In Section II we briefly describe the nuclear reaction models included in the GNASH code along with some recent improvements that have been made. Section III shows comparisons between the new

evaluated data generated with GNASH and measured data, to benchmark the evaluations. Section IV discusses extensions that have been made in MCNP to handle the new evaluated data and some preliminary results. Section V summarizes and discusses future work that is needed.

### The GNASH Code

The GNASH code calculates nuclear reaction cross sections using equilibrium and preequilibrium theories, and makes use of predetermined direct reaction cross sections calculated with codes such as ECIS and DWUCK. A capability exists to use either the exciton model or the Feshbach-Kerman-Koonin quantum theory for preequilibrium emission. However, for generating APT data libraries, the exciton model was used because of the shorter computational time needed, and FKK calculations were performed only for a selection of reactions to help validate the exciton model results. Sequential equilibrium emission is accounted for by using the Hauser-Feshbach theory with a full conservation of angular momentum and parity. All reaction chains are included providing their cross sections exceed 0.1 mb, which usually required calculating the decay of approximately 100 excited nuclides for a typical target nucleus.

Full details of the nuclear reaction theories included in the GNASH code are described elsewhere (Young et al., 1996). Here, we briefly describe some recent improvements in the code relevant for modeling reactions up to 150 MeV: multiple preequilibrium emission; calculation of nuclear recoils; and improved optical models.

We have developed (Chadwick et al., 1994) a theory of multiple preequilibrium emission that accounts for the emission of more than one fast preequilibrium ejectile. Such mechanisms are important for predicting the magnitude of the preequilibrium emission spectra while maintaining flux conservation, and they strongly influence the production cross sections of residual nuclei. Currently the model in GNASH allows for the emission of a second preequilibrium ejectile, and as will be shown later, enables measured secondary emission spectra to be described reasonably well.

Another recent improvement in the GNASH code system is a capability to calculate the energy spectra of secondary heavy nuclear recoils (Chadwick et al., 1996a). Kinematical transformations were simplified by making use of closed-form expressions that can be obtained due to the analytic nature of the Kalbach angular-distribution systematics equations. A proper treatment of the slowing-down of the compound nucleus following fast preequilibrium emission was made, along with its subsequent increase in kinetic energy (on average) during sequential evaporation. Inclusion of recoil information in the evaluated libraries

is important for accurate calculations of radiation heating (KERMA, Kinetic Energy Released in Matter) and damage.

Finally, we note that optical model analyses are extremely important for GNASH calculations since they provide transmission coefficients, the reaction cross section, and elastic scattering angular distributions. Extensive previous work in Group T-2 on optical models up to a few hundred MeV is being supplemented by a new collaboration between Los Alamos and Livermore (Madland and Dietrich, 1996) to develop an improved global optical potential for use in APT analyses.

### Comparisons of Evaluated Data with Measurements and LAHET

Evaluations up to 150 MeV for incident neutrons and protons have been completed for isotopes of Pb, W, Fe, O, and C, and in the coming months we shall produce libraries for D, N, Al, Si, Ca, Cr, and Ni isotopes. In order to validate these libraries, the evaluated data have been benchmarked extensively against existing measured data. In this section we compare our evaluated data - determined using the GNASH code - with measured data for a selection of cases that are particularly important in APT (Pb, W, Fe and C). Some comparisons with LAHET results are also made to illustrate the advantages of using evaluated data libraries up to 150 MeV.

The importance of using evaluated data libraries can be clearly seen in Figure 1, where the evaluated tungsten proton nonelastic cross section is compared with LAHET calculations and with measured data. In fact, only one tungsten measurement exists, and so other nonelastic experimental values are shown from nearby elements (tantalum) and from systematics of measurements on other targets. It is evident that LAHET agrees well with measurements above a few hundred MeV but underestimates the nonelastic cross-section at lower energies. The evaluated data library, being based on an optical model calculation, describes the measurements below 150 MeV well. This is important since secondary particle production cross sections, such as neutron production (which is so important to APT), are proportional to the nonelastic cross section.

Figures 2 - 4 show benchmark comparisons of the evaluated cross sections for lead, for incident protons and neutrons. Figure 2 shows the calculated neutron production spectra for incident protons at various angles compared with the Meier et al. (1989) 113 MeV measurements. Agreement is good, except at 150 degrees, though the discrepancies here have minimal practical importance since the back-angle cross sections are very small. In Figure 3, angle-integrated neutron and proton emission spectra are shown for 90 MeV protons incident on lead. No experimental data exist

for lead at this energy, so the calculated results are compared with data on bismuth, which would be expected to be similar. This figure demonstrates that the evaluated libraries, based on GNASH calculations, correctly account for the relative amounts of secondary neutron and proton production. In Figure 4 secondary neutron emission following 65 MeV neutrons incident on lead is compared with new forward-angle UC-Davis measurements by Hjort et al. (1996). The good agreement between evaluation and measurement is important due to lead's role as a neutron multiplication blanket in APT.

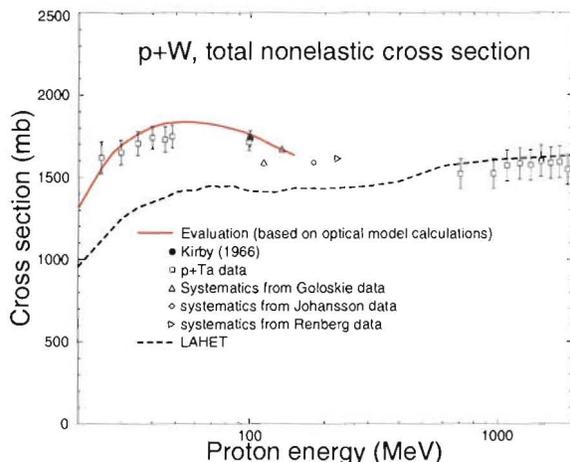


Figure 1. The new evaluated tungsten proton nonelastic cross section is compared with LAHET calculations and with experimental data from the CSISRS database at the National Nuclear Data Center (<http://www.nndc.bnl.gov/>).

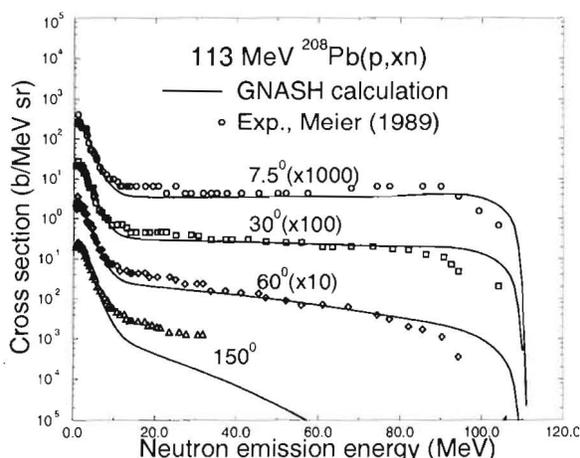


Figure 2. The GNASH calculated neutron production spectra at various angles for incident protons on  $^{208}\text{Pb}$  are compared with the Meier et al. (1989) 113 MeV measurements.

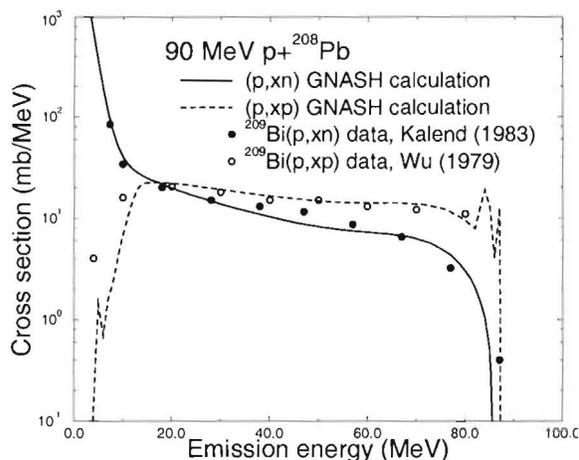


Figure 3. Angle-integrated neutron and proton emission spectra are shown for 90-MeV protons incident on Pb. No comparable experimental data exist for Pb, so experimental data for Bi are shown for comparison.

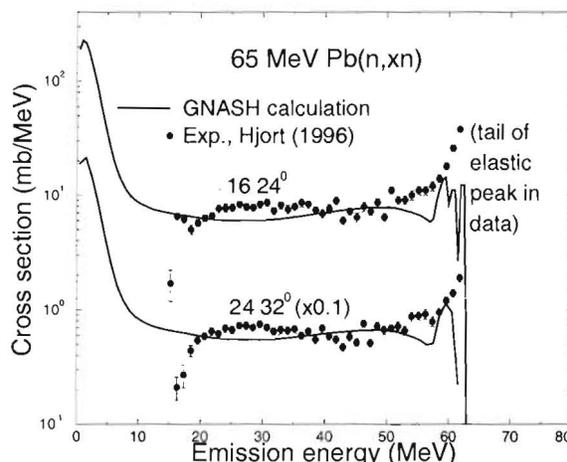


Figure 4. The GNASH calculated secondary neutron emission following 65 MeV neutrons incident on Pb is compared with new UC-Davis measurements by Hjort et al. (1996).

Figures 5 - 7 show comparisons between the evaluated libraries and measured data for tungsten, for incident protons and neutrons. Very few measurements exist for tungsten, and there is evidence suggesting that Meier et al.'s 113 MeV data is approximately 50% too high (Chadwick and Young, 1996b). First, measurements of neutron production at backward angles were made by Skyrme (1962) for 43 and 150 MeV protons, and Figure 5 shows that the agreement with our calculations is reasonably good. On the other hand, the 113 MeV data of Meier et al. exceeds both the Skyrme data and our calculations. Second, when Meier

et al.'s 113 MeV data for lead and tungsten are compared, the tungsten data are seen to exceed the lead data at all emission energies. This result is surprising since tungsten has a smaller nonelastic cross section than lead, and the average spectrum evaporation energies are similar in the two cases. Third, we have made energy balance arguments in which the average excitation energy after fast preequilibrium emission is determined, and knowing the average neutron separation energy and evaporation kinetic energy, the neutron multiplicity can be determined. Such arguments show that the neutron multiplicity corresponding to the Meier et al. data appears to be

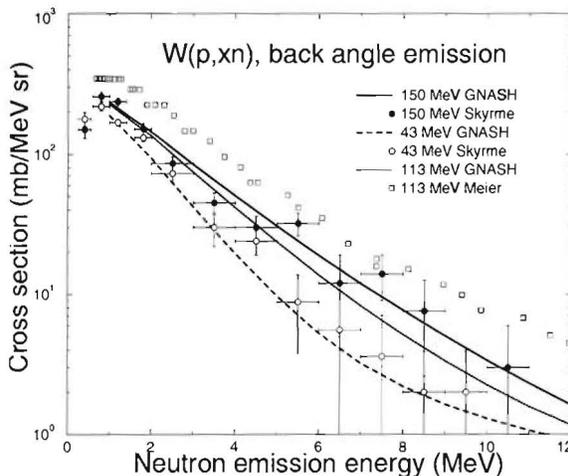


Figure 5. Comparison of back-angle GNASH calculated secondary neutron production from incident protons on W with experimental data.

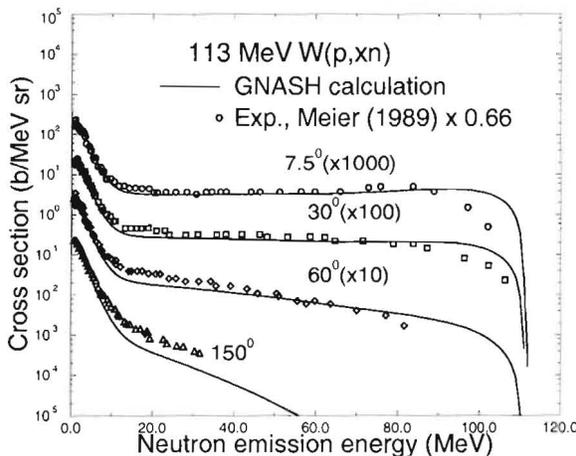


Figure 6. Comparison of GNASH calculated secondary neutron production from incident protons on W at 113 MeV with the normalized experimental data of Meier et al. (1989).

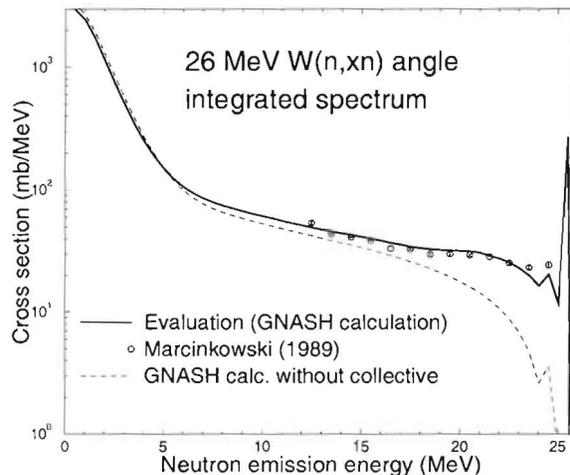


Figure 7. Comparison of GNASH calculated 26-MeV  $W(n,xn)$  angle-integrated neutron-production spectrum with measured data.

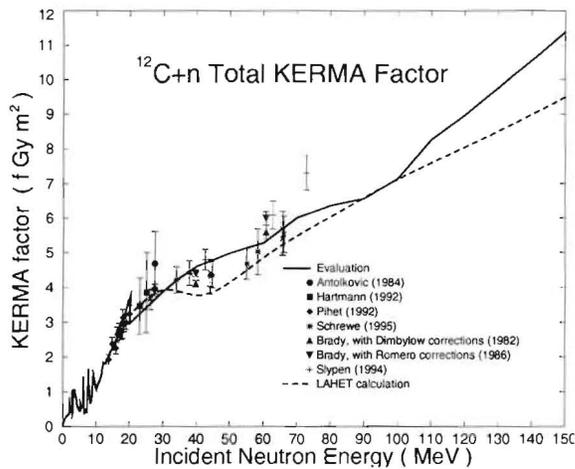
anomalously high. When the Meier et al. data is scaled down by a factor of 2/3, agreement with the evaluated data is good, as shown in Figure 6.

The only other existing emission spectra data for tungsten are the 26 MeV  $^{184}W(n,xn)$  measurements by Marcinkowski et al. (1989). Figure 7 shows that our calculated angle-integrated spectrum agrees well with the measured data, and demonstrates the importance of collective excitation mechanisms at high energies. The peak that is seen at the highest energies is due to collective excitation of the low-lying rotational levels. Additionally, collective excitation of giant isoscalar resonances was included, as described by Marcinkowski et al. (1996). The ECIS code was used to calculate these cross sections.

Finally, since carbon will be used for the APT beam stop, the calculated radiation heating due to neutrons incident on carbon is shown in Figure 8 compared with measurements. This figure shows the total KERMA factor as a function of neutron incident energy, derived from the evaluated microscopic cross sections. Note that the S.I. units of  $f \cdot \text{Gray} \cdot \text{m}^2$  can be converted to  $\text{MeV} \cdot \text{barns}/\text{atom}$  by dividing by 0.804 for carbon. The smaller KERMA factor values determined by LAHET compared to GNASH (and compared to experiment) are primarily due to LAHET's underprediction of preequilibrium deuteron emission.

### Extensions to MCNP and MCNP Data Libraries

Extensions to MCNP and its associated data libraries to fully utilize the new 150 MeV evaluations for incident neutrons have been nearly completed. In



**Figure 8. Comparison of the calculated KERMA factor with measurements for neutrons incident on C. References to the experimental data can be found in Chadwick et al. (1996c).**

this section, we summarize the changes that have been made and present some preliminary results. The current production version of MCNP (Version 4A) supports only secondary neutrons and photons from neutron collisions. The implicit assumption is that the energy from all other reaction products (light charged particles as well as heavy recoils) is deposited locally at the neutron collision site.

Only with the advent of the ENDF6 format (McLane et al., 1995) has there been a general framework available for nuclear-data evaluators to describe the angle-energy correlated spectra of reaction products. The new 150-MeV evaluations documented in the previous two sections have been prepared using this format. For incident neutrons and protons, secondary reaction product cross sections and angle-energy correlated spectra have generally been provided for neutrons, photons, protons, deuterons, tritons,  $^3\text{He}$  and alphas. Additionally, secondary energy spectra have been provided for the heavier recoil nuclei ( $A \geq 5$ ) in the evaluations, but this information is not included explicitly in the corresponding MCNP data library, but is used in calculating the appropriate heating numbers.

To accommodate the additional data provided in the new evaluations, the format for continuous-energy incident neutron MCNP data tables has been re-defined to allow for a complete representation of production data for an arbitrary number of secondary particles (Frankle, 1996). To meet these specifications, a code has been written that reads, processes, and adds all appropriate secondary charged-particle data from the evaluations to an existing MCNP data table (Little, 1996). Total production cross sections are formed for each particle, and differential secondary spectra (energy distributions, angular distributions, and correlated

energy/angle distributions) are processed as a function of incident neutron energy for each reaction producing the particle.

A modified version of MCNP has been produced that utilizes the expanded neutron data libraries. The patch that created this version enables the code to read the new data tables, manipulate the charged-particle production data as necessary, determine the number and weight of secondary charged particles to produce at each neutron collision, sample the neutron reaction that produced a specific charged particle, sample the secondary energies and angles, and write the information to the particle bank. At this time, no subsequent transport of the secondary charged particles is performed. That capability will be enabled shortly as part of the previously-mentioned merging of MCNP and LAHET functionality. Then, proton transport will be accomplished based on the new 150 MeV proton-incident evaluations and/or the existing LAHET models.

Some preliminary results have been obtained using the modified version of MCNP and the new APT data libraries, and are reported for slabs of C and Pb in Table 1. "Thin" slabs are approximately 1 mean free path (mfp) at the source energy, while "thick" slabs are approximately 5 mfp at the source energy. The total number of secondary charged particles produced is calculated for each particle type. In all cases, the neutron source is a monoenergetic 150-MeV beam incident perpendicular to the slab. Neutrons are terminated if they scatter below 20 MeV. LAHET results are presented for comparison. Table 1 indicates that total proton production is in reasonable agreement (although we have not compared secondary energy spectra here), but the production of heavier charged particles is substantially different between the two codes.

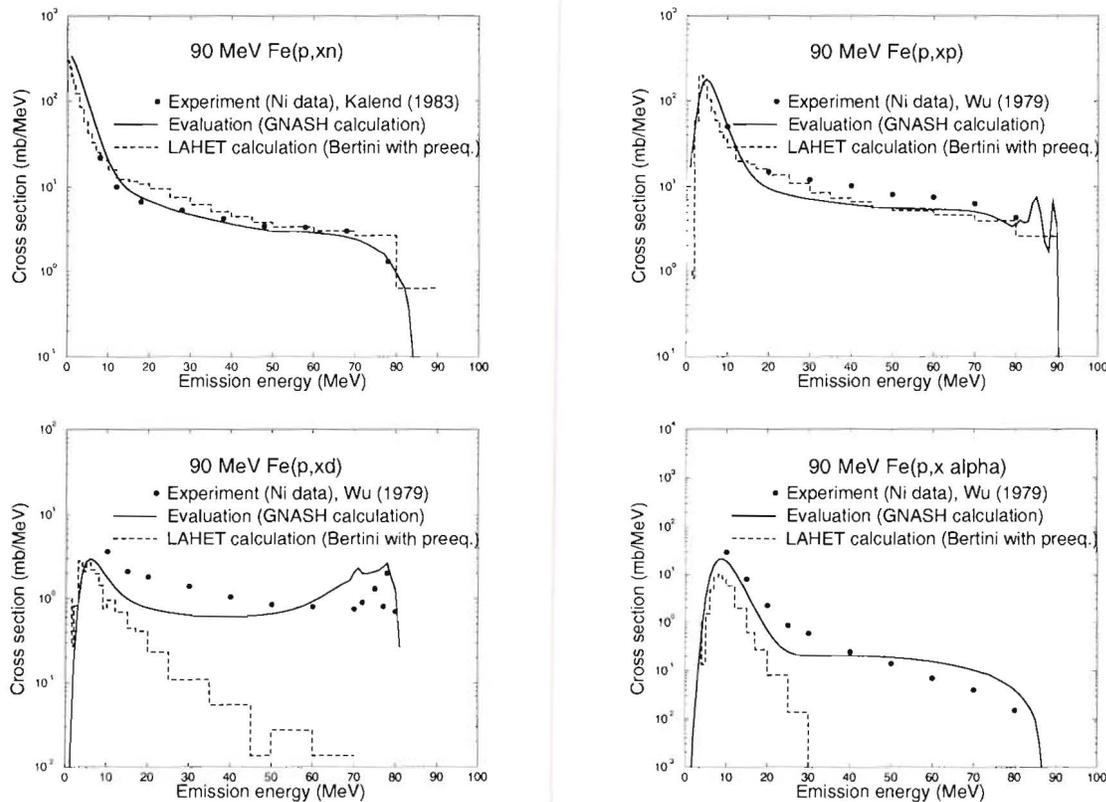
To understand the differences in particle production between LAHET and MCNP (using the new libraries), we compare in Figure 9 calculated angle-integrated particle-emission spectra following 90-MeV protons incident on Fe with measurements. As no measured data exist for Fe here, we compare against Ni data, which we expect to be similar to Fe, and which exists for n, p, d, and alpha ejectiles from University of Maryland (Kalend et al., 1989; Wu et al., 1979). It is clear that while both LAHET and GNASH calculations describe the neutron and proton spectra reasonably well, LAHET largely underpredicts d and alpha emission, particularly at high energies. This underprediction is because the Bertini INC model does not include preequilibrium cluster emission mechanisms. Differences do exist between the GNASH calculations and the measurements, but the GNASH calculations account for the main features seen in the data, particularly the preponderance of high energy clusters ejectiles. Thus, the smaller values for d, t, and

alpha emission for LAHET in Table 1 are due to LAHET's underprediction of preequilibrium cluster emission. Likewise, since LAHET does not include fast cluster emission processes, more energy is available for n and p emission, resulting in the slightly higher proton production in Table 1.

We note that while LAHET largely underpredicts fast cluster emission, the practical consequences of this underprediction for APT are probably small - the influence on tritium production estimates will be minor, though LAHET calculations of heating may be low.

**Table 1. Comparison of total secondary charged-particle production for thick and thin slabs of C and  $^{206}\text{Pb}$ .**

	Protons			Deuterons			Tritons			Alphas		
	MCNP	LCS	Ratio M/L	MCNP	LCS	Ratio M/L	MCNP	LCS	Ratio M/L	MCNP	LCS	Ratio M/L
<b>C-thin</b>	0.529	0.579	0.91	0.258	0.107	2.41	-	-	-	0.758	0.940	0.81
<b>C-thick</b>	1.326	1.535	0.86	0.649	0.301	2.16	-	-	-	2.192	2.934	0.75
<b><math>^{206}\text{Pb}</math>-thin</b>	0.215	0.257	0.84	0.0084	0.0043	1.95	0.0023	0.0014	1.64	0.0182	0.0072	2.53
<b><math>^{206}\text{Pb}</math>-thick</b>	0.652	0.795	0.82	0.026	0.0137	1.90	0.0065	0.0039	1.67	0.0511	0.0198	2.58



**Figure 9. Comparison of secondary particle production for protons incident on Fe at 90 MeV. The calculated results using GNASH and LAHET agree with measured spectra fairly well for neutron and proton production. GNASH accounts for the deuteron and alpha spectra reasonably well, though LAHET largely underpredicts these measurements.**

## Summary

To summarize, our work has emphasized greater accuracy in nuclear data over the energy range from 20-150 MeV for the APT program. Models in the nuclear theory code GNASH have been developed and improved. Evaluated nuclear data (based on GNASH calculations) for incident neutrons and protons for several important APT materials have been completed. The evaluations have been compared to available experimental data with favorable results. Based on the evaluations, MCNP data libraries for incident neutrons have been prepared and used in a modified version of MCNP that allows for an arbitrary number of secondary particles from neutron collisions. Preliminary results, when compared with corresponding LAHET calculations, indicate reasonable agreement in the total number of neutron-induced protons, but factors of 2-3 disagreement in the total number of heavier charged particles produce due to improved modeling in GNASH and the corresponding data libraries.

The APT nuclear data and particle transport efforts are ongoing projects. Future priorities include:

- Continued upgrades to GNASH with further enhancements to the multiple preequilibrium emission model, incorporation of new total and nonelastic experimental cross section data from WNR at LANSCE, and utilization of new optical models currently being developed.
- New evaluations and corresponding libraries for incident neutrons and protons on D, N, Al, Si, Ca, Cr, and Ni isotopes with further nuclides in future years.
- Upgrades to the nuclear data processing code NJOY for handling the expanded format for the neutron evaluations and libraries, and to produce libraries for incident charged particles.
- Continuation of work on merging MCNP and LAHET into one seamless code, including improvements in the treatment of angular deflections in multiple scattering theory.
- Providing multigroup n-particle libraries for possible use in deterministic shielding codes.

The goal of this work is to ensure an accurate predictive capability for (n/p), (T/p), shielding, heating, activation, and damage applications relevant to the design and operation of an APT facility.

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