Benchmarking Simulation Methods for Methane Moderators*

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The Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory is a spallation neutron source dedicated to materials research. Its three cryogenic methane moderators provide twelve neutron beams to fourteen instruments and test facilities. IPNS has begun a program to enhance the effectiveness of its target, reflector, and moderator system. This program will involve the examination of many different potential system modifications and their various interactions. The first component of such a task is the assessment of the current performance of the target, reflector, and moderator system, and the verification of the computational model used to evaluate the system. To that end, we have developed a Monte Carlo model of the IPNS neutron generation system as currently configured, and are in the process of validating that model with corresponding measurements. This paper describes the Monte Carlo model and analysis, as well as some of the experimental comparisons we use to validate our model.

1. IPNS DESCRIPTION

IPNS is a spallation neutron source in which protons are accelerated to 450 MeV and guided onto a light water-cooled target composed of depleted uranium disks clad with Zircalloy. A rough schematic of the target, reflector, and moderator system appears in Figure 1. The IPNS accelerator system delivers some 14 μA of protons in bursts less than 100 ns long at a rate of 30 Hz. The horizontal uranium target, 250 mm long and 100 mm in diameter, is surrounded by a (vertical) graphite inner reflector 250 mm in diameter, and a beryllium outer reflector 600 mm in diameter. There are three cryogenic moderators, each decoupled from the reflector by 0.5 mm of cadmium. The "F" moderator is below the front of the uranium target (the end near the proton beam), and is composed of liquid methane (CH_4) at 100 K. The F moderator is 100 by 100 by 45 mm in size, poisoned with gadolinium 16 mm below the large faces, both of which are viewed by beam ports. The "H" moderator is above the front of the uranium target, and consists of solid methane at 30 K. The H moderator is also 100 by 100 by 45 mm in size, and is poisoned with gadolinium at the centerline. For both the F and H moderators, the poisoning sheets are 0.5 mm thick gadolinium-aluminum alloy containing 17.2 weight percent gadolinium. The "C" moderator is below the rear of the uranium target, and is also solid methane. The C moderator is an un-poisoned, re-entrant moderator, 100 by 100 by 80 mm, with horizontal grooves 40 mm deep and 10 mm high in the viewed face.

2. SIMULATION METHODS

We have begun a program of computational study of various potential enhancements to the IPNS target, moderator, and reflector assembly. The first step in any such program is the validation and benchmarking of the software used to perform the calculations, as well as the employment of that software and the interpretation of its results. These simulations are being done using the MCNP [1] (version 4B) and LAHET [2] (version 2.70) computer programs. These codes are extremely popular,

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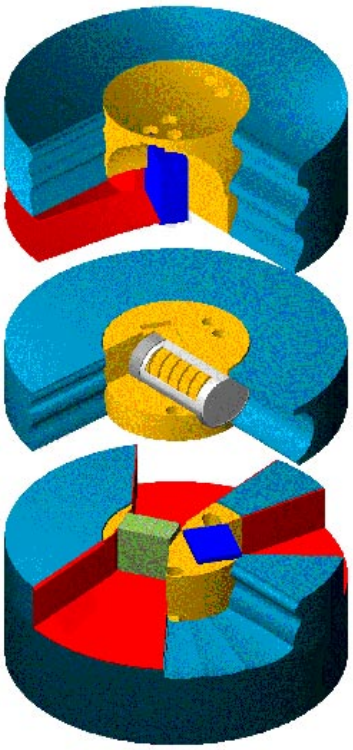

Figure 1. The IPNS target, reflector, and moderator system. Protons enter from the rear on the left in this view.

and in general quite well validated. [3] Our task is then to benchmark the application of the codes to our IPNS system (including cross section data specifically applicable to cold moderator materials), and to indentify appropriate characterizations which can be both simulated and measured.

The calculations were done on Intel-P6 based computers running the Linux operating system. The MCNP calculations were, for the most part, performed on a virtual parallel machine composed of four such computers using the PVM software from Oak Ridge National Laboratory [4]. MCNP is written directly to take advantage of PVM; no modification is required. We are currently in the process of doubling the capacity of this cluster.

MCNP, like any modern Monte Carlo code, has a rich selection of variance reduction features from which to chose. We employed the so-called *Weight Window* and *Point Detector* methods, described below, to speed the convergence of our calculations. Using variance reduction is absolutely essential for obtaining worthwhile estimators of cold neutron production in reasonable amounts of computer time. As an example, consider the calculation of the intensity of neutrons in a typical range of interest, say 4–14 Å. The use of these variance reduction methods speeds the calculation to a specified precision by a factor of 10^4. The MCNP4B describes these variance reduction methods in detail. [1] A short description of these methods appears below.

2.1 Weight Windows

Weight window variance reduction is a method of simultaneous space- and energy-dependent splitting and roulette techniques. The basic concept is that, as particles move from one region of phase space to another, more “interesting” region, they are split—i.e., replaced by multiple identical particles

with the same total weight as the initial particle. These particles are then followed individually. Conversely, in the roulette instance, particles moving from one region of phase space to a *less* interesting region have some finite probability of being terminated. If the the particle survives the roulette process (the fractional probability of which is p) then its weight is multiplied by $1/p$. This process vastly increases the fraction of CPU time spent on particles in neutronically important portions of phase space, yet does not introduce a bias into the resulting calculations. The use of weight windows requires then some assessment of the importance of each region of phase space sampled within the problem. This importance function can be estimated by a skilled user, or from an adjoint multi-group calculation. MCNP also includes a very effective method for the iterative generation of the importance function.

The phase space importance function used for these calculations was constructed using this automatic generation method (called the Weight Window Generator). The importances thus calculated are subject to statistical errors, as are all quantities calculated by Monte Carlo techniques. In order to speed the convergence of the weight window generator, intermediate results were averaged for "similar" regions of geometrical space. In other words, the weight window generator might, for example, estimate the neutronic importance of several different cells, each of which was physically part of the cadmium decoupler surrounding a given moderator. When these importances are averaged, the statistical variation is smoothed somewhat, and the iterative process converges more quickly.

Finally, the resulting importance function was smoothed, in order to guarantee no severe gradients in the importance function. During the iterative process, the repeated appearance of such severe gradients in a given location indicates that the adjacent regions of phase space are "too large." Breaking these regions into smaller ones, either in physical space or in energy space, results in a better importance function and a more efficient calculation.

Using a well-defined importance function can result in interesting regions of phase space (such as a viewed moderator surface between 1 meV and 1 eV) being sampled hundreds of times more frequently than they would be in a strictly analog simulation, with corresponding increases in overall computing speed.

2.2 Point Detectors

The point detector method of variance reduction is not actually a biasing of the random walks in the Monte Carlo calculation, but is instead a next event estimator. One of the great disadvantages of Monte Carlo methods is that they are intrinsically unsuited to calculating a particle flux at a given point, as opposed to averaging over a large volume. Since the production of neutron beams is essentially an intentional streaming problem, a strictly analog Monte Carlo calculation would almost never track particles in the small portion of phase space that is of interest.

If one is interested in the flux at a given point in space, one can instead deterministically calculate the probability that a particle at any other given point in space will contribute to the flux at that first point. In MCNP, this is implemented as the point detector. At every scattering event, the transmission probability from the scattering event location to the detector location is calculated, and the point detector tally is augmented by the product of that probability and the particle's weight. Thus every single collision which takes place anywhere in the system contributes to the calculation, as opposed to only the miniscule fraction of the particles that happens to leak through the system in the correct direction.

Consider a perfectly moderating material—i.e., all scattering is inelastic, and there is no absorption. A source neutron entering this moderator has perhaps 5 MeV of energy, and will eventually leak out of the moderator. If one considers only the viewed face of the moderator, the chance that the neutron leaks out of the appropriate face is approximately 25%. Thus a tally recording all leakage through the viewed moderator face records some 0.25 contributions per source particle.

In a hydrogenous material, a 5 MeV neutron requires about twenty inelastic collisions to reach

energies of 1 meV. Using a point detector, every one of those collisions contributes to the detector tally. Even when one considers only those scattering events below, say, 10 eV, there are a minimum of eight to ten collisions that result in desirable neutrons contributing to the tally. Once elastic collisions are included, each source particle can contribute to the tally up to twenty times or more. Thus the efficiency of the calculation per source particle increases by nearly two orders of magnitude. This increase comes at a relatively trivial cost of some ten percent or less in CPU time per source particle. This hundred-fold increase in calculation speed, when multiplied by the hundred-fold increase described above from the weight window methods, can turn a calculation that might take the better part of a year into one which will be completed in an hour with the same statistical precision.

3. COMPARED QUANTITIES

The quantities examined in these simulations include the neutron energy-dependent intensity of the neutron beams, and the neutron energy-dependent emission time distributions of the neutron beams. While these do not provide a complete description of the moderator's performance, such as might be required for the optimized design of a scattering instrument, these two functions of neutron energy do encompass the most significant characteristics of a neutron beam.

3.1 Intensity

The intensity of the neutron beams emitted from the viewed moderator faces, normalized by the accelerator beam current, corresponds to what in optics terminology is a normalized "luminous intensity." This intensity is related to a measured flux;

$$i(E) = \frac{L^2}{I}\,\phi(E)|_L\,, \tag{1}$$

where $\phi(E)$ is the time-averaged flux per unit energy at a distance L far from the moderator face, and I is the time-averaged accelerator beam current. In general, the use of the luminous intensity rather than flux as a metric permits the brightness of the moderator face to be specified independently from the length of the flight path. For this metric to be useful for a re-entrant moderator such as the IPNS "C" moderator, the distance L must be either large enough that there is no significant self-shielding of the moderator face, or L must be similar to the actual distance from the real moderator to the real sample. The units of $i(E)$ are then neutrons per steradian per second per electron-Volt per micro-ampere. The commonly-quoted measure of moderator coupling effectiveness is then

$$[E \times i(E)]|_{E=1\ \mathrm{eV}}\,. \tag{2}$$

If the intensity $i(E)$ is multiplied by E, the resulting quantity is propportional to the normalized counting rate seen in a thin $1/v$ detector placed at a distance L from the moderator surface, scaled by a factor which is independent of neutron energy. Thus $E \times i(E)$ is easily compared to a time-of-flight beam intensity measurement. This quantity is calculated directly in the Monte Carlo simulations, and benefits from both the point detector variance reduction method (which results in *every* scattering event in *every* history contributing to the tally) and the weight windows variance reduction method (which results in more scattering events in neutronically relevant portions of phase space).

3.2 Pulse Shapes

The time-dependence of the neutron emission from the moderator surface is a strong determinant of the resolution which can be achieved in any experiment involving the neutron beam. These pulse shapes have been measured, as a function of neutron energy, using a time-focused crystal analyzer system described elsewhere. [5, 6] We are presently repeating and extending these measurements, but the

experimental results reported here are those previously reported. [6] The quantity measured in such an experiment is simply a counting rate as a function of time. A crystal reflects a series of mono-energetic pulses to a detector. The operation of the time-focused crystal analyzer is such that the only instrumental contribution to the resolution of the measurement is the flight time of the neutrons across a 1 mm thick crystal and their lifetime in a 1 mm thick detector. Thus, the peak shape of the counts registered in the detector matches the time distribution of the neutrons leaving the moderator surface, delayed by the known flight time of each order of reflection.

In the Monte Carlo calculations, this quantity is determined by tallying the neutrons leaving the viewed surface of the moderator as a function of time from the initial source pulse. The difference between the simulation and measurement is then limited to the minimal instrumental resolution and flight time of the neutrons from the moderator to the detector. Unfortunately, this tally does not benefit from the use of the point detector method described above, although it does benefit from the weight windows. The point detector tally could be configured to tally pulse shapes as well, but the memory requirements to record pulses on a microsecond time-scale extending over a range of milliseconds would be impractically large.

4. SCATTERING KERNEL DATA

The accuracy of any Monte Carlo simulation is limited by the accuracy of the scattering kernel data used in the simulation. This is especially true in the case of under-moderated, time-dependent systems such as those we are studying. The scattering kernels of greatest importance are those for the cryogenic moderator materials. The kernels that are widely available for use with MCNP appear in Table 1, and are a part of the standard MCNP distribution. The kernels are described in greater detail elsewhere. [7]

Table 1. Cryogenic moderator material scattering kernels available for use with MCNP.

Material	(K)	Material	(K)	Material	(K)
Solid Methane	22	Para Hydrogen	20	Para Deuterium	20
Liquid Methane	100	Ortho Hydrogen	20	Ortho Deuterium	20

5. RESULTS

We have compared the output of our Monte Carlo simulations to corresponding measurements made on the IPNS system. We have examined results from all three of the existing moderators, and we have examined both spectral intensities and wavelength/energy dependent pulse shapes. We have examined moderators composed of both liquid and solid methane, moderators with and without heterogeneous poisoning, and moderators of reentrant and non-reentrant geometries.

5.1 Spectral Intensities

We have measured the absolute spectral intensities of the neutron beams on several IPNS beam-lines. Here we will discuss only the measurements on two of these beam-lines—the QENS beam-line, viewing the solid methane “H” moderator, and the SEPD beam-line, viewing one side of the liquid methane “F” moderator. A more complete description of these measurements appears elsewhere in these proceedings. [8]

In the case of the solid methane moderator, the scattering kernel information used in the simulations corresponds to a temperature of 22 K, rather than the 30 K at which our solid methane moderators

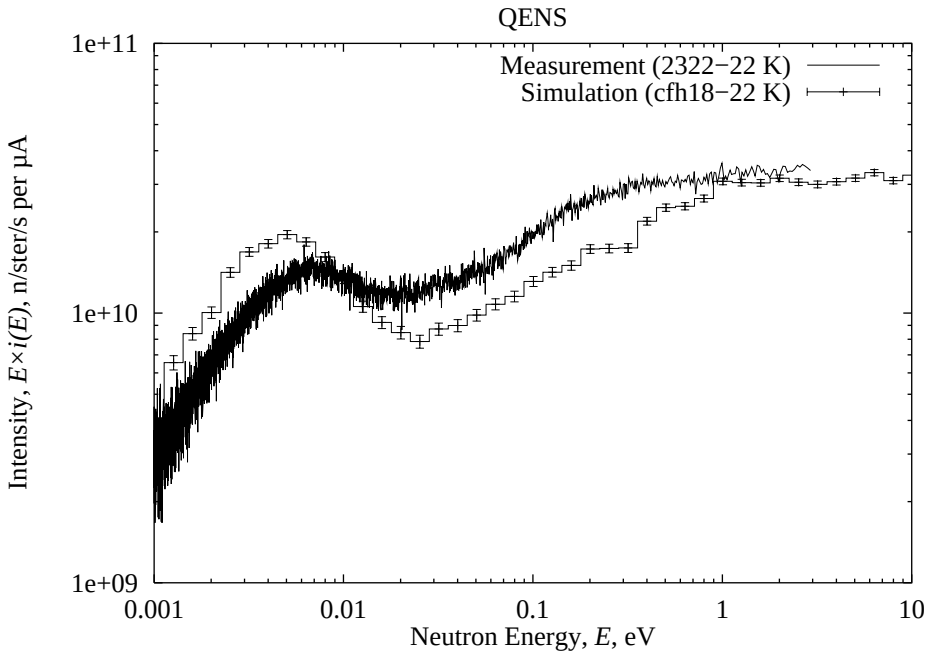

Figure 2. Absolute spectral intensity as measured on QENS from a 22 K solid methane moderator.

normally operate. With this in mind, we operated the solid methane "H" moderator at 22 K for a short time during a dedicated machine research period. The absolute spectral intensity for the QENS beam line (normal to the moderator surface), as measured at 22 K using the efficiency determined with a gold foil activation run at 30 K, is shown in Figure 2, together with the corresponding simulation results. This simulation tracked approximately 10^8 neutrons, of which some forty percent were original (spallation) source neutrons, as produced by the LAHET code following 5×10^4 450 MeV proton cascades. The point detector tally used recorded contributions not only from collisions within the moderator volume, but also from the portions of the graphite reflector which are nominally viewed through the beam line collimation, albeit perhaps after transmission through moderator and decoupler regions.

The intensity was measured over a period of approximately one hour, at an average proton current of 12.36 μA. The measured moderator coupling is 3.3×10^{10} neutrons per steradian per second per micro-Ampere of proton current. This quantity represents the integral over the entire 100 mm by 100 mm viewed surface of the moderator. The measured moderator coupling matches the coupling predicted by simulation to within the experimental and Monte Carlo precisions at 1 eV, but the shapes match poorly, as we discuss below. The nominal time-averaged moderator brightness at 1 eV, assuming constant proton operation at 14 μA, is 4.6×10^{13} neutrons per steradian per second per square meter.

The liquid methane "F" moderator is typically operated at 100 K, matching the temperature in the scattering kernel library. We measured the spectral intensity for the liquid methane moderator on the SEPD beam-line, normal to one side of the "F" moderator. Figure 3 shows the absolute spectral intensity as measured, again with the corresponding simulation results. These Monte Carlo results came from the same simulation as described above for the QENS measurement—tracking approximately 10^8 neutrons, as produced by the LAHET code following 5×10^4 450 MeV proton cascades and subsequent multiplication by both variance reduction methods and physical processes. This point detector tally also recorded contributions from viewed portions of the reflector as well as from the moderator volume,

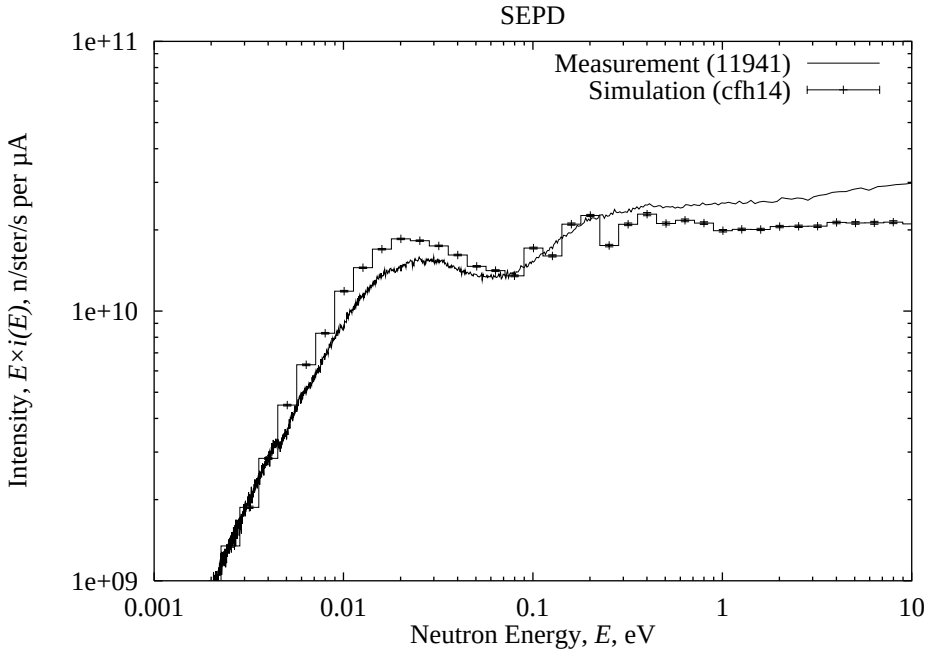

Figure 3. Absolute spectral intensity as measured on SEPD from a 100 K liquid methane moderator.

although the "F" moderator is distinct in that there is no reflector region immediately "behind" the moderator, as the moderator is viewed from both sides.

The intensity was measured over a period of some ninety minutes, at an average proton current of 13.95 μA. The measured moderator coupling is 2.4×10^{10} neutrons per steradian per second per micro-Ampere of proton current. As before, this quantity represents the integral over the entire 100 mm by 100 mm viewed surface of the moderator. The simulated moderator coupling and subsequent average brightness is lower than that measured by approximately 30% (1.7×10^{10} and 2.3×10^{13}, respectively). The nominal moderator brightness at 1 eV, assuming constant proton operation at 14 μA, is 3.3×10^{13} neutrons per steradian per second per square meter.

5.2 Pulse Shapes

The pulse shapes of the liquid methane "F" moderator and of the grooved solid methane "C" moderator have been previously measured. [6]. These measurements were performed using a time-focused crystal analyzer based on the (nnn) series of reflections from a cooled germanium crystal. The pulse shapes were only measured for neutron energies corresponding to these reflections at the specific Bragg angle, in this case $2\theta = 120^\circ$. Table 2 lists these reflections.

The fourth order reflection (40.49 meV neutron energy) as measured on the liquid methane "F" moderator appears in Figure 4, together with the corresponding simulation results. Space limitations prohibit the inclusion of all of the twenty pulse shapes measured. However, one significant metric describing the neutron pulses is the pulse width. The measured pulse width as a function of neutron wavelength appears in Figure 5, and is compared to the simulated value. Note that Figure 5 does not show the often quoted full-width at half-maximum of the pulse, but rather the time width of the central eighty percent of the integrated pulse area. This metric is somewhat more robust than the FWHM for noisy pulses—the counting statistics on the measured pulses are rather poor at both extremes of the

Table 2. The allowed (nnn) reflections from germanium at 25 K and $2\theta = 120°$.

n	λ (Å)	E (meV)	v (km/s)	n	λ (Å)	E (meV)	v (km/s)
1	5.686	2.531	0.6958	12	0.4738	364.4	8.350
3	1.895	22.78	2.087	13	0.4374	427.7	9.046
4	1.421	40.49	2.783	15	0.3791	569.4	10.44
5	1.137	63.27	3.479	16	0.3554	647.8	11.13
7	0.8123	124.0	4.871	17	0.3345	731.3	11.83
8	0.7107	162.0	5.567	19	0.2993	913.6	13.22
9	0.6318	205.0	6.262	20	0.2843	1012	13.92
11	0.5169	306.2	7.654	21	0.2708	1116	14.61

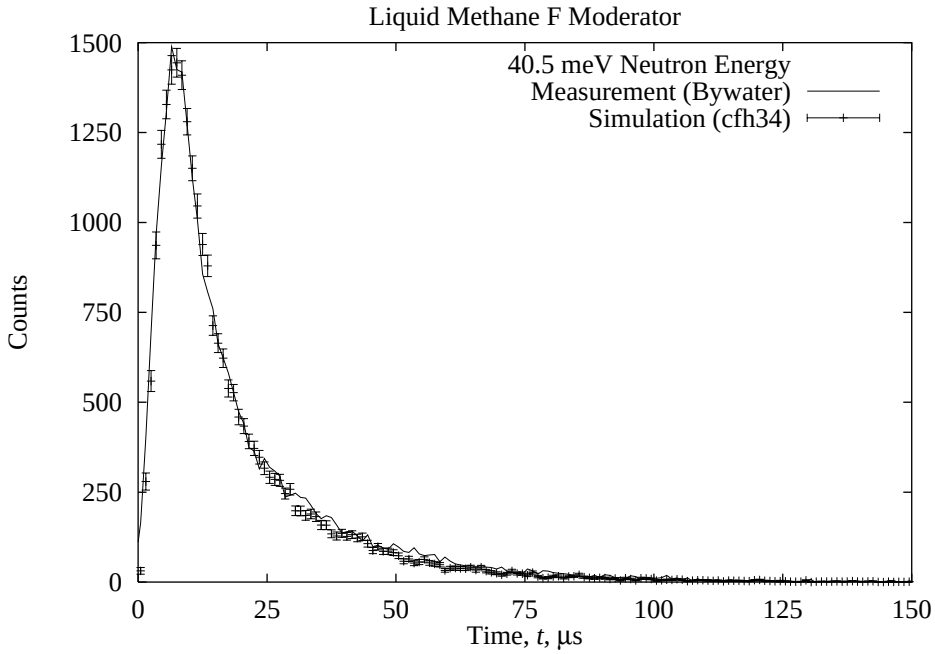

Figure 4. Time-dependent pulse shape for Ge (444) neutrons from a 100 K liquid methane moderator. The line represents the measured data, while the points are the simulation results.

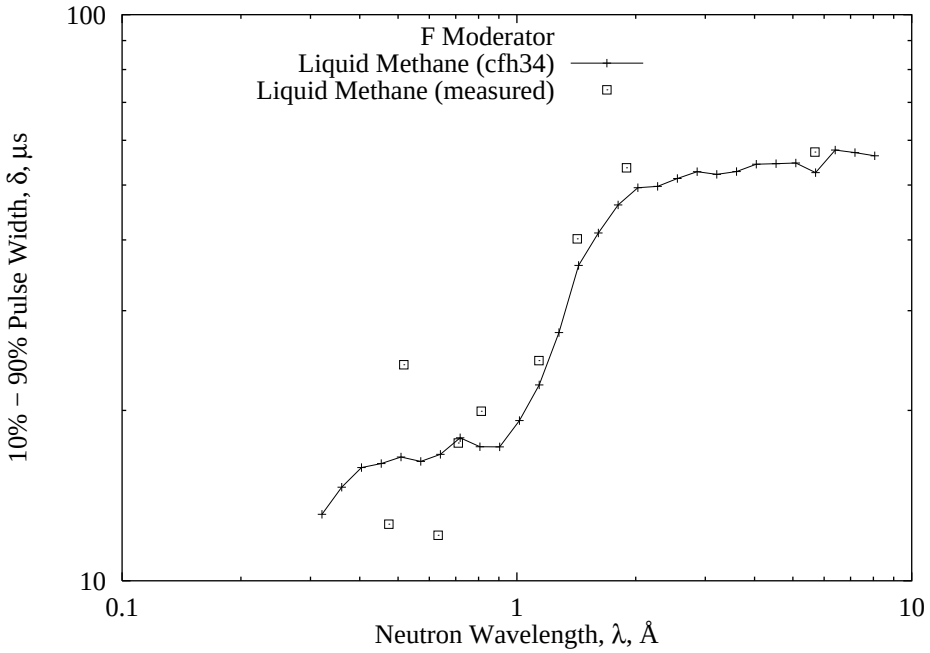

Figure 5. Pulse width as a function of neutron wavelength for the liquid methane "F" moderator.

measured range, and the Monte Carlo precision is rather poor at the long-wavelength end of the range. This metric is also more sensitive to the tails of the pulses, which are poorly sampled in the simulation.

The 40.49 meV pulse shape for the solid methane "C" moderator (note that the pulse shapes for the solid methane "H" moderator have not yet been measured) appear in Figure 6, from both measurement and simulation. The measured pulse width for the "C" moderator as a function of neutron wavelength appears in Figure 7, and is compared to the simulated value.

6. DISCUSSION

The results of our simulations do, to a large extent, behave in a fashion similar to the corresponding measured characteristics. There are, as discussed below, serious difficulties with the neutron moderation predicted in solid methane, but these difficulties are not so pervasive that we cannot use the simulation model for the optimization and analysis studies which we wish to perform. That is, we think that the calculated changes will be reliable enough to support many optimization decisions.

6.1 Spectral Intensities

Figure 2 compares the results of simulation and measurement for the spectral intensity of the solid methane "H" moderator. While the spectrum displays many qualitative similarities, there are obvious, considerable discrepancies. The moderator coupling (the intensity at 1 eV) is nearly identical. This would seem to indicate that the portion of the calculation above 1 eV is very accurate. It should be remembered that errors in the absolute scaling of the measured spectrum would result in a uniform shift in the intensity, independent of neutron energy. Thus, any measurement errors should lead to a constant factor being multiplied by the intensity over all energies.

However, at energies below 1 eV, the simulation predicts significantly different intensities than those actually measured. This discrepancy is so pervasive that the spectral temperature of the moderator

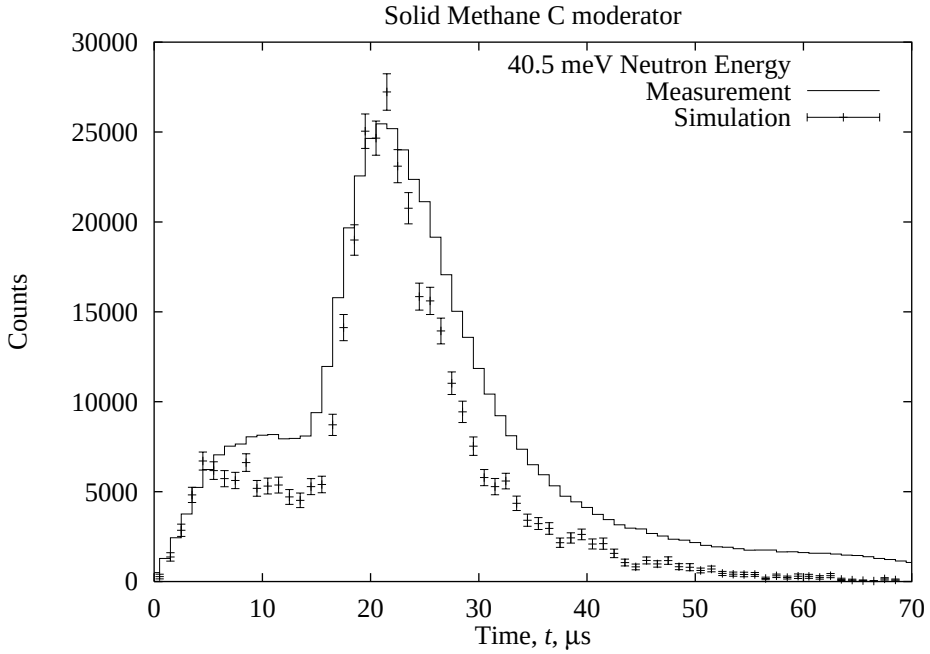

Figure 6. Time-dependent pulse shape for Ge (444) neutrons from a 30 K solid methane grooved moderator.

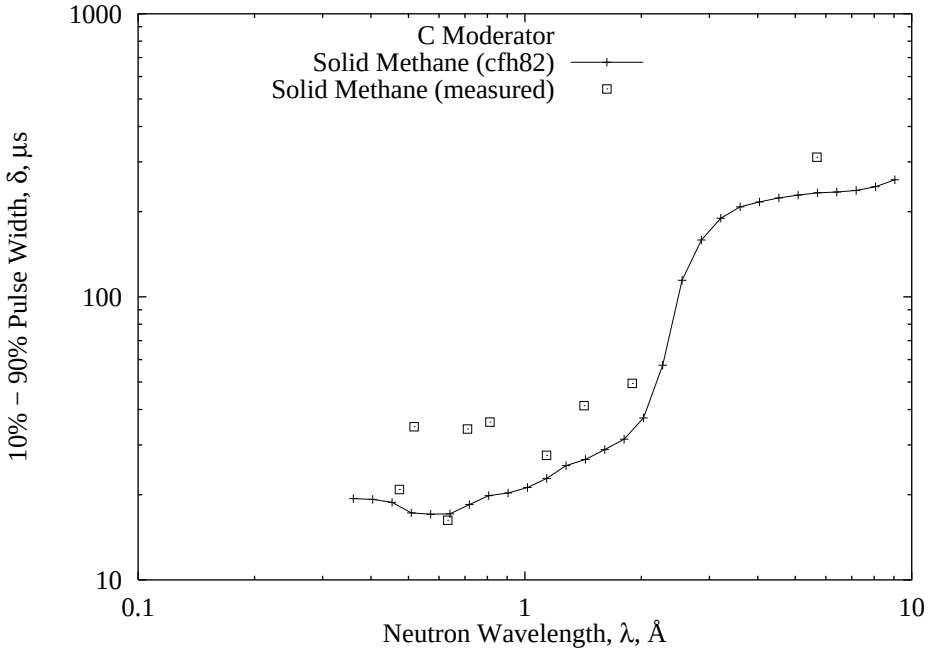

Figure 7. Pulse width as a function of neutron wavelength for the grooved solid methane “C” moderator.

is incorrectly estimated, as well as the actual thermal flux, as might be characterized by the thermal-to-epithermal ratio. The measured spectral temperature is approximately 4.2 meV (49 K), while the simulated spectral temperature is about 2.5 meV (29 K). Note that the maximum value of a Maxwellian in $i(E)$ when expressed as $E \times i(E)$ occurs at $2k_{\mathrm{B}}T$. Furthermore, the cutoff energy, defined as that energy where the flux changes from a $1/E$ behavior to a Maxwellian behavior, occurs at a significantly different energy in the simulation from that actually observed. As a result, the simulation significantly under-predicts moderator performance from 10 meV to 1000 meV, the range most relevant to, say, powder diffraction and chopper spectrometer measurements, and significantly over-predicts moderator performance for cold neutrons, as relevant to quasi-elastic scattering and small-angle diffraction.

Finally, the simulated spectrum has several non-physical features, including several discontinuities, below 1 eV. It is worth noting that the simulation treats the molecular scattering kernels in detail at neutron energies of 950 meV and below. The onset of inaccuracies and non-physical features just below 1 eV seems indicative of some problems with the scattering kernel data, coming from either inaccuracy in the molecular description, or from discretization error in that description. The kernel is stored, in the code, as a set of equally likely final energies for each initial energy, and a set of equally likely scattering angles for each pair of energies. For this solid methane kernel, there are sixteen final energies for each incident energy, and sixty-four scattering angles for each pair of energies. The non-physical features observed are much less than those in the liquid methane kernel, where there are only eight final energies for each initial energy, and eight scattering angles for each pair of energies. This could conceivably result in considerable discretization error, as is described below.

The accuracy of the simulation of the liquid methane spectrum, as in Figure 3, is considerably better. There are slight deviations between measurement and simulation in the thermal region, perhaps on the order of 30%. The moderator coupling also shows an error of about 30%, but in the other direction from the deviations at thermal energies. However, the spectral temperature is predicted fairly accurately. The largest difficulty with the liquid methane simulation is, again, the non-physical features (especially around 200 meV) which we believe to be caused by discretization errors in the scattering kernel representation.

When taken together, the two spectra indicate a possible problem in the geometric model of the system, given that the moderator coupling is accurately predicted in the case of the solid methane moderator, but not in the case of the liquid methane moderator. At and above 1 eV, the cross section information should be equally valid, so any discrepancy which appeared only in one instance would be more likely to come from the geometric model than the physics of the simulation. We have not yet uncovered the origin of this problem.

6.2 Pulse Shapes

The simulated pulse shapes from the liquid methane moderator match the measured pulse shapes nearly perfectly, as exemplified in Figure 4. Similarly, the pulse widths as shown in Figure 5 seem to behave very similarly.

Again, the solid methane simulations were not so successful. There are significant differences in the relative heights of the the bimodal peak from the grooved moderator, as seen in Figure 6. The under-prediction of the height of the earlier peak (from the tips of the moderator fins) also results in an under-prediction of total pulse width. Examination of Figure 7 clearly shows that the solid methane simulation consistently under-predicts observed pulse widths by 30% to 40% over all wavelengths. On a more positive note, the rise time of the pulse appears to be accurately predicted. This is quite important for many instruments in which the resolving power of the instrument is largely determined by the sharpness of that rise time, as opposed to the total width of the pulse.

It is further obvious that the selection of neutron wavelengths at which the pulse shape was

measured is not well matched to the physics of the solid methane system. The sharp increase in pulse width, corresponding to the increasing importance of the storage term in the pulse shape as the neutrons come into thermal equilibrium with the moderator material, is very inconveniently located in the exact range that is not measured at all in the germanium (nnn) series of reflections with $2\theta = 120^\circ$. These measurements should be repeated for a set of wavelengths that will better match the characteristics of the solid methane moderators.

7. CONCLUSIONS

The Monte Carlo simulation of the methane moderators at IPNS shows clear difficulties with the solid methane scattering kernel as implemented with the MCNP computer code. These problems take the form of significant errors in the spectral temperature, intensity, and pulse shapes of the moderated neutron flux. These errors are not present in the liquid methane simulations, which match measurements quite well.

While the solid methane discrepancies are significant enough to warrant serious attention, we do believe that the comparison between the measurements and the simulations is good enough that we can consider our simulation model benchmarked for aspects of the target, moderator, and reflector system that do not rely upon the solid methane scattering kernels—e.g., target design, reflector material, moderator position. For studies involving detailed study of moderation in solid methane, however, our model requires further development and testing.

One avenue for such testing involves comparison of the scattering kernel used in the simulations to a scattering kernel actually measured on an inelastic neutron spectrometer. We propose that such measurements be undertaken over a wide range of Q- and ω-space for solid methane at the temperature used in the development of the simulation data. Such a program of measurements could further be extended to cover novel moderator materials, as well as conventional moderator materials at a wider range of temperature and pressure conditions than has been considered previously.

Finally, the intensity and pulse shape measurements used for these comparisons should be extended. While the intensity measurements appear to be satisfactory, the pulse shape measurements, using a crystal analyzer arrangement, do not have sufficiently dense wavelength coverage. The germanium series used reflects an adequate selection of wavelengths for the liquid methane moderators, but not for the solid methane moderators, as no reflections exist in the critical region between two and five Ångstroms where the pulse width changes so rapidly.

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