MEASUREMENT AND FITTING OF SPECTRUM AND PULSE SHAPES OF A LIQUID METHANE MODERATOR AT IPNS

J. M. Carpenter

Argonne National Laboratory
Argonne, Illinois 60439

R. A. Robinson

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

and

A. D. Taylor
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

and

Rutherford Appleton Laboratory
Oxfordshire, England

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Abstract

We have measured the absolute intensity, and the energy spectrum, and the pulse shapes as function of neutron energy for the IPNS liquid CH_4 "F" moderator, at 108 K. We have fitted the spectrum, corrected for attenuation by aluminum in the beam, using a new cut-off function and fitted the pulse shapes to a new function which is the sum of two decaying exponentials, convoluted with a gaussian, and determined the wavelength variation of the parameters. We present here the results of a preliminary analysis.

Introduction

Information on the spectrum, the absolute intensity and pulse shapes as function of energy, for pulsed source moderators is essential for the design of neutron spectrometers and for the analysis of measurements. Liquid methane, CH_4 is of special interest because of its high proton density, its excellent thermalization properties, and its good heat removal capacity in flowing systems. During the time that the cryogenic moderator-reflector system was operating at IPNS, we measured the neutronic properties of the liquid methane "F" moderator. Table I gives the parameters of the "F" moderator. Figure 1 shows the cryogenic moderator-reflector system.

Fig. 1 The IPNS cryogenic moderator-reflector system. The liquid-methane "F" moderator is at the bottom, viewed from both sides. The text gives the physical parameters of the moderator.

Table 1

Dimensions:	10.2 x 10.2 x 5.1 cm ²
Heterogeneous poisoning:	none
Decoupler:	1 x 10 ²⁰ Gd/cm ²
Void liner:	4 x 10 ²¹ 10B/cm ²
Reflector:	30 cm ϕ Be @ 100 K, 60 cm ϕ Be ambient
Aluminum in beam path:	1.11 cm ~ 100K, .52 cm ambient
Temperature:	108 K

We used the resonance activation of a .027-mm gold foil¹ to determine the absolute epithermal beam current, with the result,

$$I_{\text{epi}} = EI(E) \quad 1\text{eV} = 2.91 \times 10^{10} \text{ n/sr} \cdot \mu\text{A} \cdot \text{s}$$

for 451 MeV protons on the depleted-uranium target. This compares quite favorably with the result calculated by our HETC-VIM monte-carlo simulations²

$$I_{\text{epi}} = 2.57 \times 10^{10} \text{ n/sr} \cdot \mu\text{A} \cdot \text{s}$$

(scaled from 2.95 x 10¹⁰ at the conditions of the calculation, 500 MeV, to 451 MeV according to the relative neutron yield, $Y(E) \propto (E_{\text{GeV}} - .12)^3$)

A thin BF_3 "pancake" beam monitor detector placed 13.69 m from the moderator surface provided the wavelength distribution of the neutrons. We fitted a function of the form

$$C(\lambda) = \eta(\lambda) \left[a_1 \frac{\lambda_T^4}{\lambda^5} \exp(-\lambda_T^2/\lambda^2) + a_2 \Delta(\lambda) \lambda^{2\alpha-1} \right] T(\lambda)$$

to the counting rate per unit wavelength. Here, we took the efficiency $\eta(\lambda) \propto \lambda$, a_1 represents the magnitude of the maxwellian term, λ_T is the characteristic maxwellian wavelength, a_2 is the magnitude of the slowing down term, $\Delta(\lambda)$ is the "joining function" which we took to have the novel form devised by Taylor⁴

$$\Delta(\lambda) = [1 + \exp(a_3 \lambda - a_4)]^{-1}$$

α is the leakage exponent, and $T(\lambda)$ a transmission correction for the structural aluminum in the beam. We find Taylor's joining function to be superior to any of those suggested (for other purposes) by Wescott.

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The transmission correction $T(\lambda)$ was taken to be of the form

$$T(\lambda) = \exp(-n \sigma_T(\lambda))$$

$$\sigma_I(\lambda) = \begin{cases} \sigma_{lo}(\lambda/\lambda_{co}); & \lambda > \lambda_{co} \\ \sigma_{Bragg}(\lambda) + \sigma_{inel}(\lambda); & \lambda_{co} < \lambda < .95 \text{ \AA} \\ \sigma_o = 1.49 \text{ b} & \lambda > .95 \text{ \AA} \end{cases}$$

The low energy cross section, $\sigma_{lo} = .77 \text{ b}$ which represents sub-Bragg inelastic scattering, was taken from the "Barn book" for $\lambda > \lambda_{co} = 4.67 \text{ \AA}$. The structured part of the cross section was modeled⁶ assuming perfect polycrystalline FCC aluminum with Debye-Waller factors taken for 300K and a Debye spectrum with $\theta_D = 36.92 \text{ meV}$,

$$\sigma_{Bragg}(\lambda) = (\sigma_o/V_o) \frac{\lambda^2}{4} \sum_{\tau < 2k} \frac{Z(\tau)-2W(\tau)}{\tau} e^{-2W(\tau)}$$

where $k = 2\pi/\lambda$, τ are the reciprocal lattice vectors $\tau = 2\pi/d$, and $Z(\tau)$ are the reciprocal lattice weighting factors and V_o the unit cell volume. The inelastic term was modeled assuming

$$\sigma_{inel}(\lambda) = \sigma_o (1 - e^{-2W(2k)});$$

(we recognize that this approximation is not correct). The resulting fitted parameters (a_1 and a_2 are unnormalized as tabulated below) were

$$\begin{aligned} a_1 &= 7.61 \\ a_2 &= 2.34 \\ T &= 11.1 \text{ meV } (\lambda_T = 2.71 \text{ \AA}) \\ \alpha &= .06 \\ a_3 &= 5.95 \times 10^{-1}, a_4 = 6.65 \end{aligned}$$

Fig. 2 Pulse widths at fraction f of maximum Δt_f , and the absolute beam current per unit lethargy, $Ei(E)$, for the L-CH₄ "F" moderator. The current is for 451 MeV protons on the depleted-uranium target. The extrapolation of pulse widths to higher energies is according to the high-energy limiting values for an infinite medium of liquid methane.

Figure 2 shows the fitted spectrum, plotted as the beam current per unit lethargy, $Ei(E)$ normalized to the measured epithermal beam intensity at 1 eV. The corrected spectrum was not free of Bragg-edge structure, even though we adjusted the aluminum thickness in attempts to produce a smoother spectrum. We believe this indicates a preferred orientation in the grain structure of the aluminum.

Using roughly $2.5 \times 2.5 \text{ cm}^2$ crystals of mica ($d = 9.94 \text{ \AA}$) and Ge(110) ($d = 2.00 \text{ \AA}$) and one detector of the SEPD at $2\theta = 152.3^\circ$, we measured the shape of the neutron pulses at wavelengths $n\lambda = 2d \sin \theta$ for the first six orders of reflection from Germanium, and for the third through twenty-third orders of reflection from artificial mica. The total flight path length from moderator to detector was approximately 15.5 m. (The distance was not well-enough known to enable independent determination of emission time delays.) Figure 3 shows the peaks measured for three typical wavelengths. The data have not been corrected for resolution of the instrument.

Fig. 3 Pulse shapes for three wavelengths, measured with a mica crystal. Times are in microseconds.

We chose to fit a function which is the sum of two decaying exponential functions, convoluted with the same gaussian. Although there are excellent theoretical grounds for fitting a sum of chi-square distribution functions to the data (at shortest wavelengths, the shape is expected to be of the form $(t-t_0)^2 \exp-a(t-t_0)^2$; Coceva⁶ has recently fitted Monte Carlo simulations of the ORELA target response to such a function), the purpose of our fitting is to provide functions useful in Rietveld profile analysis. Existing Rietveld codes use a function which is the sum of a rising exponential, joined continuously to a falling exponential, convoluted with a gaussian. This function is non-physical (neutrons emerge before the source pulse) but provides a reasonably good fit to room-temperature polyethylene moderator pulse shapes. We found that such a function cannot adequately describe our L-CH₄ data. Use of the chosen function would require only minor modifications of the pulse shape description in existing Rietveld codes, since the pulse shape function is of mathematically similar form to that in current use.

The fitted function tends to undershoot the peaks at longest wavelengths, but is otherwise an excellent fit for the entire range of times and wavelengths.

We did not presuppose the wavelength variation of the fitted parameters except that of the longest-lived exponential, which we assumed constant, as is theoretically defensible. Figures 4, 5, and 6 give the wavelength variation of the long-lived (β_1) and short-lived (β_2) exponentials, the ratio of their amplitudes, and the gaussian standard deviation. The solid lines on Fig. 3 represent the fitted function. Resolution broadening appears as a contribution to σ , which is proportional to wavelength.

Fig. 4 Wavelength variation of the exponential-decay factors for the pulse-shape function. The value of β_1 was determined from the long-time part of the lowest-energy peaks, and held fixed in fitting the remaining parameters. The curve is a "best guess" at the smooth variation of $1/\beta_2$.

Fig. 5 Wavelength variation of the standard deviation of σ the gaussian function. The curve is a "best guess" at the smooth variation of $\sigma(\lambda)$.

Fig. 6 Wavelength variation of the fraction of the total intensity in the slow exponential. The curve is a "best guess" at the smooth variation of the fraction.

Figure 2 shows also the pulse full-widths Δt_f at fraction f of maximum, uncorrected for resolution, which we estimate to be a small effect.

We have compared our measurements to TIMOC calculations of Picton, et al.⁷ on a 90 K CH₄ moderator of the same dimensions as ours. The calculated pulse widths Δt_f are significantly smaller than our measurements, but the long-time decay at low energy agrees quite well; the calculation giving 77 μ s e-folding time, and the data 72 μ s.

Conclusion

We have measured the spectrum and pulse shapes as function of wavelength for a 108 K Liquid CH₄ moderator, and developed simple mathematical functions which provide satisfactory fits to the data.

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