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Experimental studies on neutronics of CH₃D and HD cold neutron moderators

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1. Introduction

Liquid and solid CH₄ have been used as high efficiency cooled and cold moderators in
spallation neutron sources, since CH₄ has a rotational level of about 1 meV, which is very
effective at thermalizing the neutrons to low temperatures. Moreover, CH₄ also has high
hydrogen number density. Heavy hydrogen-substituted methane, CH₃D, has lower rotational
energy levels than CH₄ but has lower number density of light hydrogen, that is, the
macroscopic scattering cross section is lower than that of CH₄. Liquid H₂ is the most
promising candidate as a cold moderator for high power spallation neutron sources, since it is
stable against radiation damage effects, but it lacks effective low energy levels to thermalize
neutrons. Molecular rotational energy levels are lower in HD molecules than in H₂ but the
neutron scattering cross section is again much lower. Therefore, both deuterated materials
have merits (lower rotational energy levels) and demerits (lower scattering cross sections),
when compared with fully protonated materials.

We do not have cross section data for these materials to study the neutronic characteristics by
simulation calculation. Therefore we have studied experimentally the neutronics of CH₃D and
HD moderators. We measured energy spectra and pulse shapes of these moderators, and we
compare and discuss the neutronic characteristics with those of CH₄ or H₂.
2. Experimental setup

We performed our experiments using the Hokkaido linac; figure 1 shows the experimental setup of a target-moderator-reflector assembly. The size of the moderator is $12 \times 12 \times 4.5$ cm$^3$. We used a graphite reflector of about 1m$^3$ volume. All moderators measured were decoupled from the reflector by 0.5mm thick cadmium plates. Energy spectra were measured by time-offlight (TOF). Pulse shapes were measured by Bragg reflection from pyrolytic graphite (PG) at a Bragg angle of 85°. The detectors are $^3$He proportional counters. The purity of D in CH$_3$D is 98% and in HD 97%. The purity of the compounds is 98%. The HD and H$_2$ moderators had been held at 18 K for about 2 hours before measurement.

Under the above conditions we measured CH$_3$D and CH$_4$ at 100K and 18K each, HD and H$_2$ at 18K.

![Fig. 1 Experimental setup](image)

Fig. 1 Experimental setup
3. Energy spectra

3.1 Comparison of CH$_3$D and CH$_4$

Figure 2 shows CH$_4$ and CH$_3$D energy spectra and the intensity ratio of CH$_3$D to CH$_4$ at 100K. Concerning the peak energies of the spectra, that is, the spectral temperatures, the peak energy is 10.8 meV for CH$_4$ and 10.6 meV for CH$_3$D and little difference was observed. From this result, we conclude that there is almost no improvement in thermalization of neutrons in CH$_3$D compared with CH$_4$. This is expected in view of the observation that the lowest energy levels of CH$_3$D and CH$_4$ are both much lower than the mean spectral energy at the temperature of 100K. The intensity of CH$_3$D is lower by 25% compared to CH$_4$ below about 50 meV. The ratio of proton number densities is approximately 3/4. So the loss of one proton per molecule is the cause of the intensity decrease. The intensity loss appears below about 40-50 meV, which is about 4 times the peak energies.

Figure 3 shows the energy spectra and intensity ratio at 18K. The peak energy of CH$_4$ is 2.51 meV and that of CH$_3$D is 2.57 meV. Below 0.01 eV, the intensity of CH$_3$D is lower by 30% compared to CH$_4$. As at 100K, at 18K the loss of proton number density more significantly influences the spectral intensity than the lowering of the energy level of molecular rotation. The intensity loss appears below about 10 meV, which is about 4-5 times of the peak energies. A characteristic feature of the difference between the spectral intensities is that the intensity is almost the same above about 4 times the peak energy. This suggests that the intensity loss takes place mainly during the diffusion process. The intensity of CH$_3$D at higher is similar.

![Energy spectra and intensity ratio of CH$_3$D to CH$_4$](image)

**Fig. 2**
Energy spectra and intensity ratio of CH$_3$D to CH$_4$

**Fig. 3**
3.2 Calculation of the spectral intensity

It is possible that the defect of low proton number density may be compensated for by using a thicker moderator. In order to study the effect of the thickness on the spectral intensity it is easier to perform simulation calculations than to perform experiments. However, there is no cross-section data for CH\textsubscript{3}D available now, but the experimental results of the CH\textsubscript{3}D and CH\textsubscript{4} spectra suggest that loss of proton number density more strongly influences spectral features of CH\textsubscript{3}D and CH\textsubscript{4} than the lower energy levels. So, we adopt the cross-section data of CH\textsubscript{4} as those of CH\textsubscript{3}D, however, changing the proton number density to a value three-fourths that of CH\textsubscript{4}. The calculation geometry is the same as that of the experiment except for the refrigerator above the moderator. We used MCNP4B to calculate the thicker version of the CH\textsubscript{3}D moderator at 100K.

Figure 4 shows the measured and calculated intensity ratios of CH\textsubscript{3}D to CH\textsubscript{4}. Because the tendencies of both plots are similar, we are led to conclude that the method is valid to estimate the thicker moderator of CH\textsubscript{3}D. Figure 5 shows integral ratios of intensities of the different thicknesses of CH\textsubscript{3}D moderators to that for 45mm thickness of CH\textsubscript{4}. From this figure, the maximum intensity ratio appears at 70mm thickness, although the intensity of the 70mm thick CH\textsubscript{3}D moderator is inferior to that of 45mm thickness of CH\textsubscript{4}. Thus, we conclude that the intensity from CH\textsubscript{3}D cannot exceed that from CH\textsubscript{4} at 100K temperature.

![Graph 1](image1.png)  
**Fig. 4** The measured and calculated intensity ratios of CH\textsubscript{3}D to CH\textsubscript{4}

![Graph 2](image2.png)  
**Fig. 5** The integral ratios of intensities of the different thicknesses of CH\textsubscript{3}D moderators to that for 45mm thickness of CH\textsubscript{4}
3.3 Comparison with HD and H₂

Figure 6 shows H₂ and HD energy spectra and the intensity ratio of HD to H₂ at 18K. Unlike the CH₄ and CH₃D spectra, the spectral shape of HD differs from that of H₂. There is the transition from para to ortho around 15meV in H₂, but there is no exactly corresponding effect in HD. The peak energy in the spectrum of H₂ is 3.04meV and that in the HD spectrum is 2.38meV: the peak energy of HD is lower than that of H₂. HD appears to thermalize neutrons significantly better than H₂, although the penalty of reduced proton density causes the intensity of HD to decrease compared with H₂. In particular, the intensity of the HD moderator is lower than in H₂ by 70% around 15meV, namely at the energy corresponding to the ortho-para transition. However, the intensity of the HD moderator approaches that of H₂ at lower energies.

![Transition from ortho to para](image)

**Fig. 6 Energy spectra and intensity ratio of CH₃D to CH₄**

4. Pulse shape and pulse widths (FWHM)

4.1 Comparison with CH₃D and CH₄
Figure 7 shows pulse shapes of CH₂D and CH₄ at 100K. The intensity of CH₂D is lower than that of CH₄. However, pulse decay times are almost the same. For 1.83meV pulses, for example, the decay time is 81.0 µs for CH₄ and 81.3 µs for CH₂D.

Figure 9 shows pulse widths (FWHM) of CH₂D and CH₄. The pulses of CH₂D turn out to be broader than those of CH₄ both at 100K and 18K.

From the pulse shape point of view, there seems to be no advantage of the CH₂D moderator compared to CH₄.

Fig. 7 Pulse shapes of CH₃D and CH₄ at 100K
4.2 Comparison with H₂ and HD

Figure 8 shows pulses of H₂ and HD at 18K. At 1.83meV, the decay time is 91.0μs for H₂ and 117μs for HD. At that energy, the decay of pulses from HD is slower than those from H₂. However, at 16.5meV (far above the energy kₚT) the decay time is 77.0μs for H₂ and 23.5μs for HD. At the higher energy, the decay of pulses from HD is much faster than from H₂. At the intermediate energy of 7.33meV, pulses from H₂ have a decay time of 81.9μs, while the pulse from HD exhibits a bimodal decay, the faster decay time being 43.0μs, the slower decay time, 102μs. We cannot claim at this time to understand these interesting observations. (The earlier discussion should be removed as above because it needs more thought.) Figure 10 shows the FWHM pulse widths of HD and H₂ as functions of the energy. Although the pulse widths of HD and H₂ are almost the same around the transition energy of 15meV, at other energies, the pulse widths of HD are broader than those of H₂.
Fig. 8 Pulse shapes of HD and H2 at 18K

Fig.10 Pulse widths (FWHM) of HD and H2
5. Conclusion

From consideration on spectra and pulses, CH$_4$D offers no visible advantages compared to CH$_3$. HD appears to thermalize neutrons better than H$_3$, although the penalty of reduced proton density gives inferior neutron characteristics at the cold neutron region. As future work, it may be worthwhile to measure a thicker HD moderator, to make up for the loss in proton density, and in order to get higher neutron intensity in the very cold neutron region.