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18.14 Experimental studies on neutronics of CH_3D and HD cold neutron moderators

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1. Introduction

Liquid and solid CH_4 have been used as high efficiency cooled and cold moderators in spallation neutron sources, since CH_4 has a rotational level of about 1 meV, which is very effective at thermalizing the neutrons to low temperatures. Moreover, CH_4 also has high hydrogen number density. Heavy hydrogen-substituted methane, CH_3D, has lower rotational energy levels than CH_4 but has lower number density of light hydrogen, that is, the macroscopic scattering cross section is lower than that of CH_4. Liquid H_2 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 H_2 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 CH_3D and HD moderators. We measured energy spectra and pulse shapes of these moderators, and we compare and discuss the neutronic characteristics with those of CH_4 or H_2.

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-of-flight (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_3D 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_3D and CH_4 at 100K and 18K each, HD and H_2 at 18K.

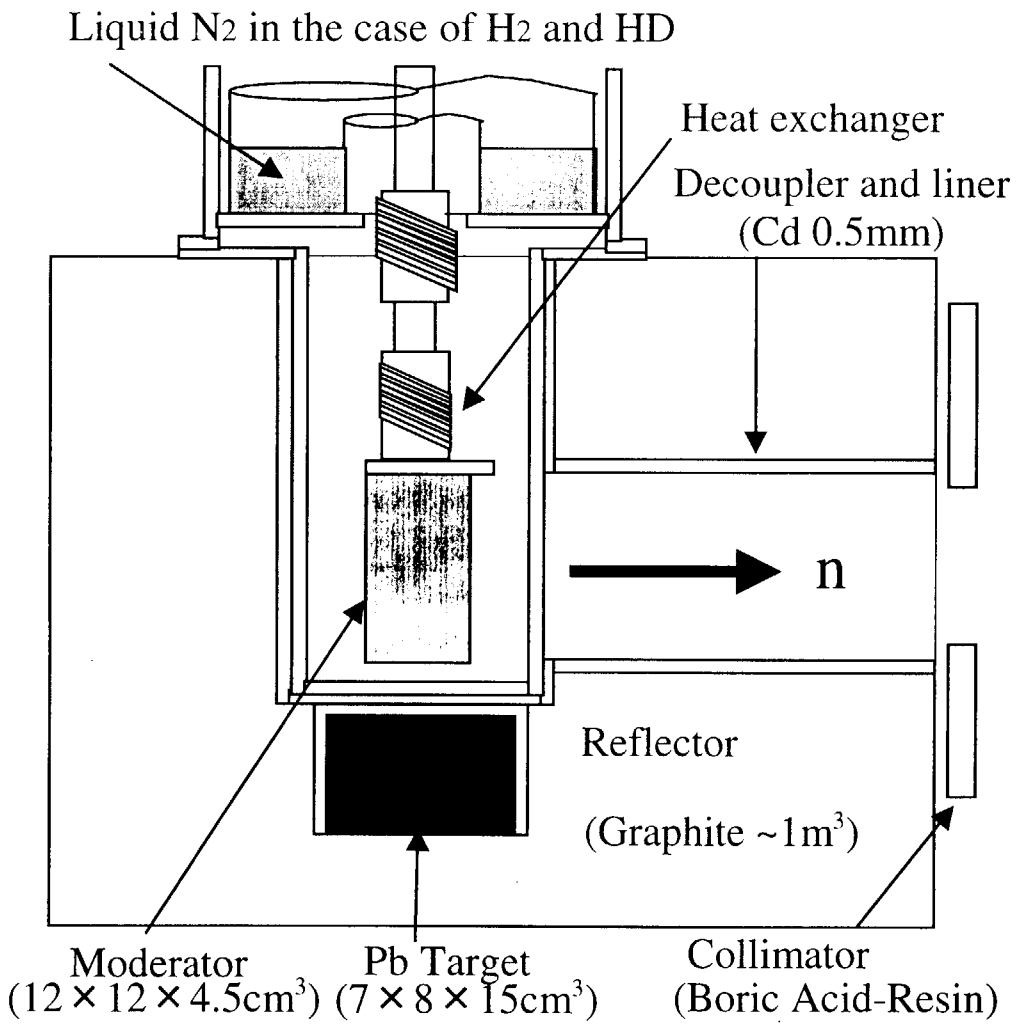


Fig. 1 Experimental setup

3. Energy spectra

3.1 Comparison of CH_3D and CH_4

Figure 2 shows CH_4 and CH_3D energy spectra and the intensity ratio of CH_3D to CH_4 at 100K. Concerning the peak energies of the spectra, that is, the spectral temperatures, the peak energy is 10.8meV for CH_4 and 10.6meV for CH_3D and little difference was observed. From this result, we conclude that there is almost no improvement in thermalization of neutrons in CH_3D compared with CH_4. This is expected in view of the observation that the lowest energy levels of CH_3D and CH_4 are both much lower than the mean spectral energy at the temperature of 100K. The intensity of CH_3D is lower by 25% compared to CH_4 below about 50meV. 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 2.51meV and that of CH_3D is 2.57meV. Below 0.01eV, the intensity of CH_3D 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 10meV, 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_3D at higher is similar .

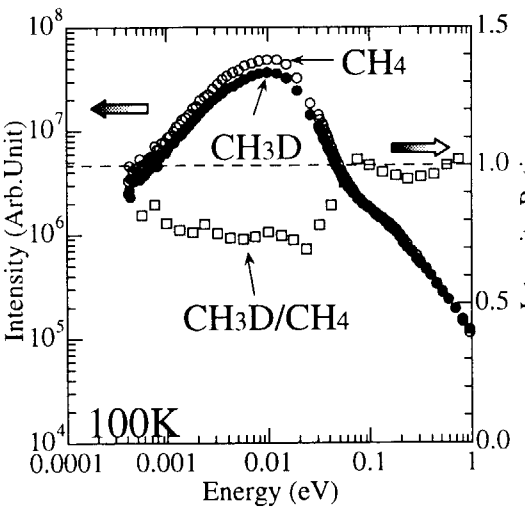


Fig. 2

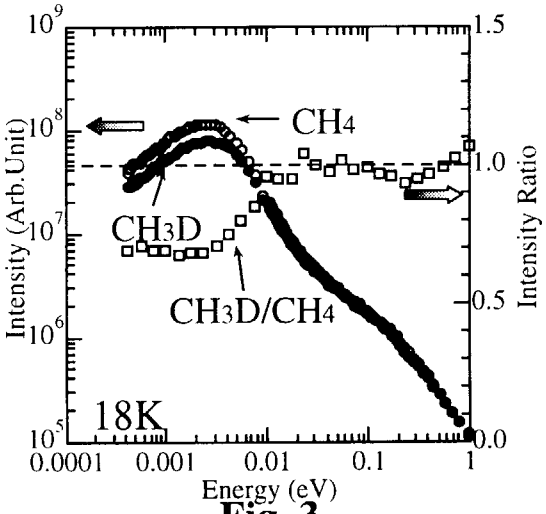


Fig. 3

Energy spectra and intensity ratio of CH3D to CH4

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_3D available now, but the experimental results of the CH_3D and CH_4 spectra suggest that loss of proton number density more strongly influences spectral features of CH_3D and CH_4 than the lower energy levels. So, we adopt the cross-section data of CH_4 as those of CH_3D, however, changing the proton number density to a value three-fourths that of CH_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_3D moderator at 100K.
Figure 4 shows the measured and calculated intensity ratios of CH_3D to CH_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_3D. Figure 5 shows integral ratios of intensities of the different thicknesses of CH_3D moderators to that for 45mm thickness of CH_4. From this figure, the maximum intensity ratio appears at 70mm thickness, although the intensity of the 70mm thick CH_3D moderator is inferior to that of 45mm thickness of CH_4. Thus, we conclude that the intensity from CH_3D cannot exceed that from CH_4 at 100K temperature.

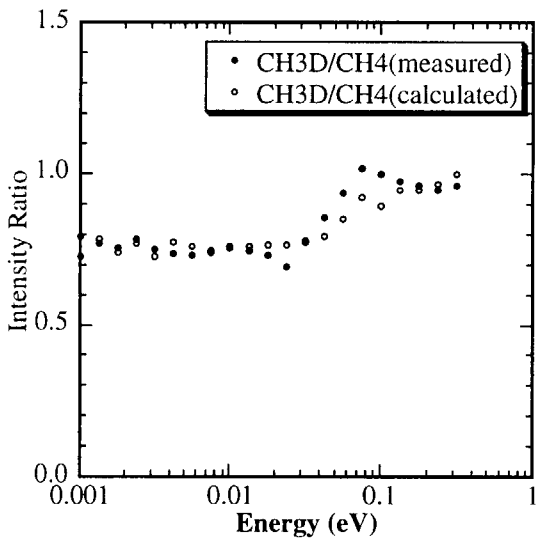


Fig. 4 The measured and calculated intensity ratios of CH_3D to CH_4

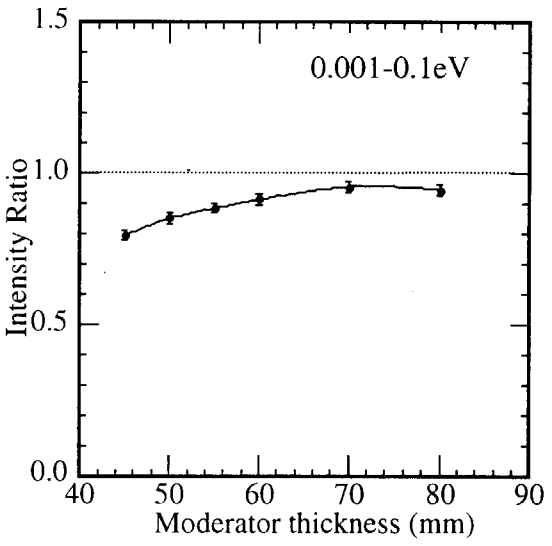


Fig. 5 The integral ratios of intensities of the different thicknesses of CH_3D moderators to that for 45mm thickness of CH_4

3.3 Comparison with HD and H_2

Figure 6 shows H_2 and HD energy spectra and the intensity ratio of HD to H_2 at 18K. Unlike the CH_4 and CH_3D spectra, the spectral shape of HD differs from that of H_2. There is the transition from para to ortho around 15meV in H_2, but there is no exactly corresponding effect in HD. The peak energy in the spectrum of H_2 is 3.04meV and that in the HD spectrum is 2.38meV: the peak energy of HD is lower than that of H_2. HD appears to thermalize neutrons significantly better than H_2, although the penalty of reduced proton density causes the intensity of HD to decrease compared with H_2. In particular, the intensity of the HD moderator is lower than in H_2 by 70% around 15meV, namely at the energy corresponding to the ortho-para transition. However, the intensity of the HD moderator approaches that of H_2 at lower energies.

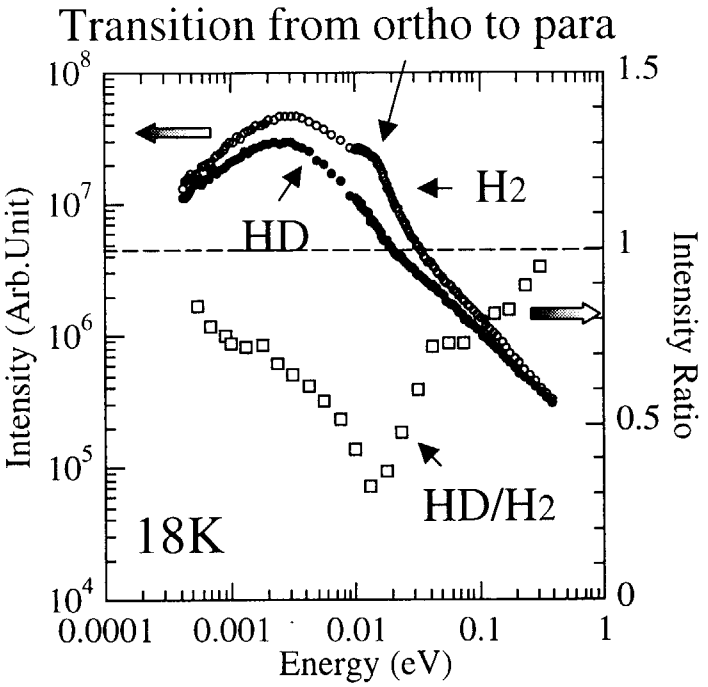


Fig. 6 Energy spectra and intensity ratio of CH_3D to CH_4

4. Pulse shape and pulse widths (FWHM)

4.1 Comparison with CH_3D and CH_4

Figure 7 shows pulse shapes of CH_4 and CH_3D at 100K. The intensity of CH_3D is lower than that of CH_4. However, pulse decay times are almost the same. For 1.83meV pulses, for example, the decay time is 81.0 μ s for CH_4 and 81.3 μ s for CH_3D.

Figure 9 shows pulse widths (FWHM) of CH_3D and CH_4. The pulses of CH_3D turn out to be broader than those of CH_4 both at 100K and 18K.

From the pulse shape point of view, there seems to be no advantage of the CH_3D moderator compared to CH_4.

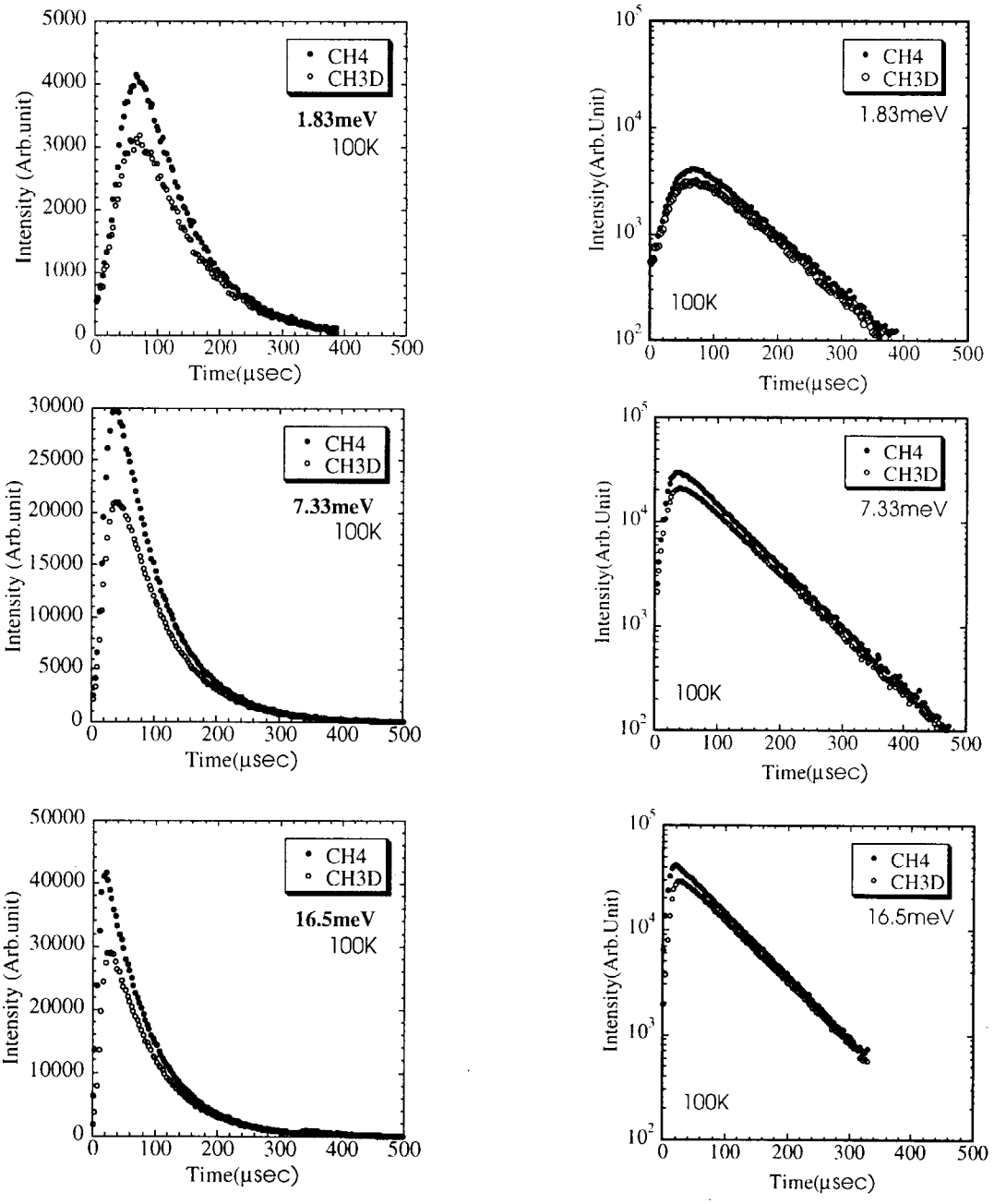


Fig. 7 Pulse shapes of CH3D and CH4 at 100K

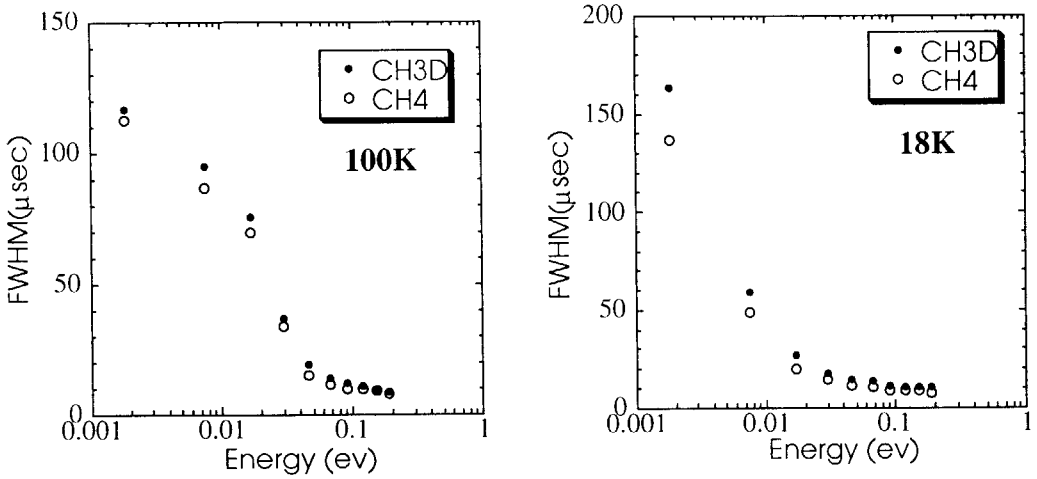
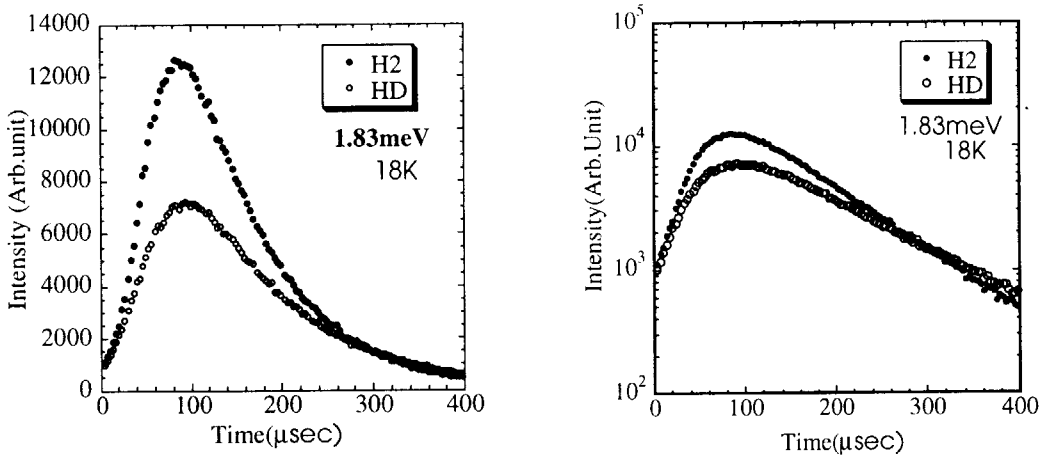


Fig.9 Pulse widths (FWHM) of CH_3D and CH_4

4.2 Comparison with H_2 and HD

Figure 8 shows pulses of H_2 and HD at 18K. At 1.83meV, the decay time is 91.0 μ s for H_2 and 117 μ s for HD. At that energy, the decay of pulses from HD is slower than those from H_2. However, at 16.5meV (far above the energy k_BT) the decay time is 77.0 μ s for H_2 and 23.5 μ s for HD. At the higher energy, the decay of pulses from HD is much faster than from H_2. At the intermediate energy of 7.33meV, pulses from H_2 have a decay time of 81.9 μ s, while the pulse from HD exhibits a bimodal decay, the faster decay time being 43.0 μ s, the slower decay time, 102 μ s. 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 H_2 as functions of the energy. Although the pulse widths of HD and H_2 are almost the same around the transition energy of 15meV, at other energies, the pulse widths of HD are broader than those of H_2.



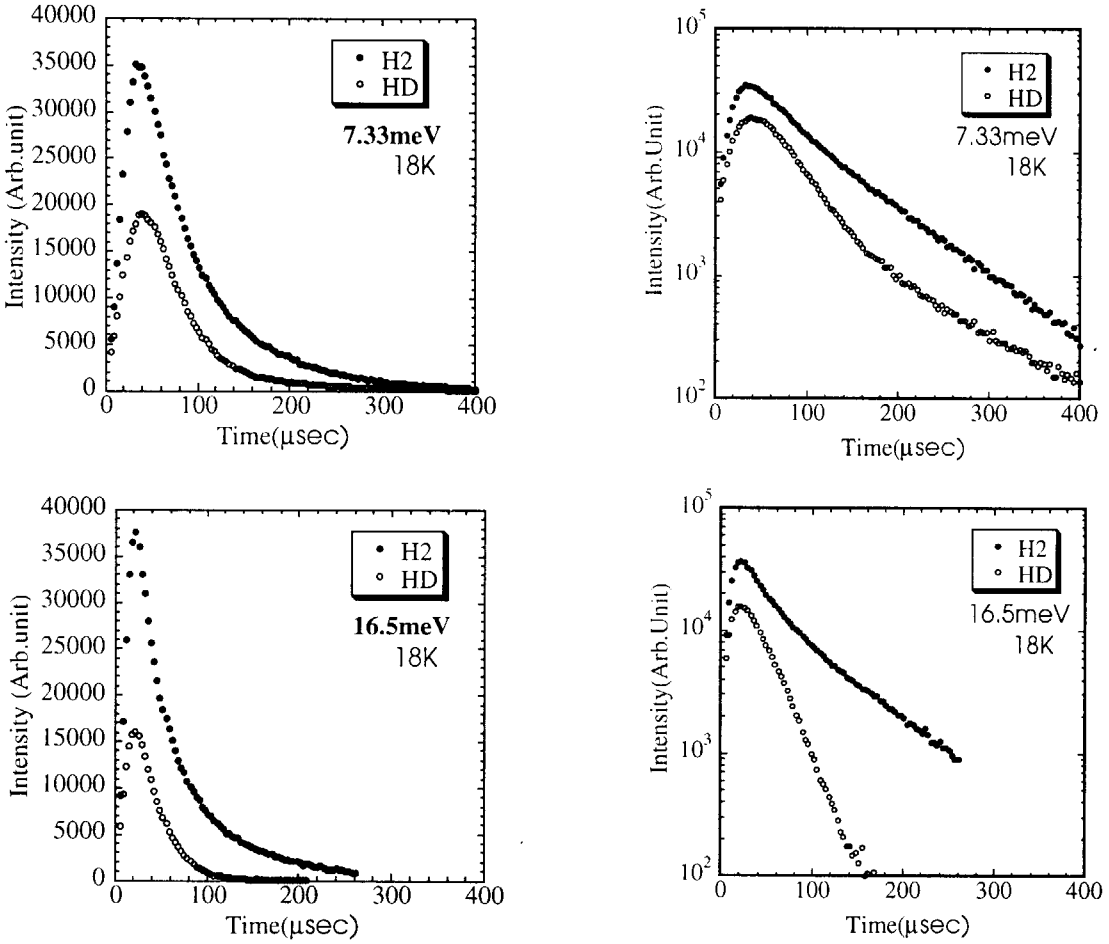


Fig. 8 Pulse shapes of HD and H_2 at 18K

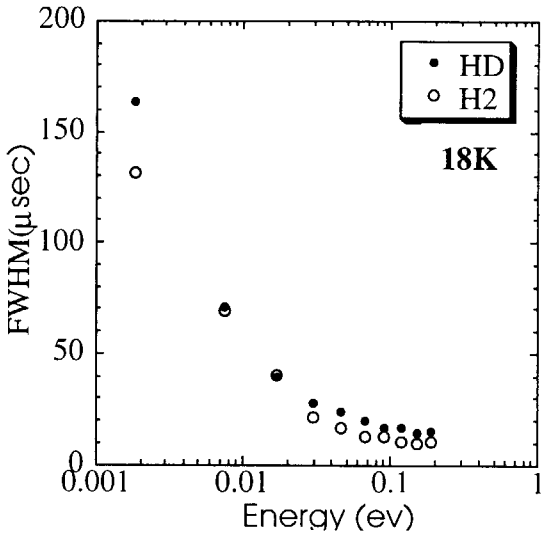


Fig.10 Pulse widths (FWHM) of HD and H_2

5. Conclusion

From consideration on spectra and pulses, CH_3D offers no visible advantages compared to CH_4. HD appears to thermalize neutrons better than H_2, 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.