

**Comparison of Decoupled Cryogenic Moderators for High Resolution Experiments**

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**Abstract**

As a cold/thermal neutron source for high-resolution experiments, we studied the neutronic performance of a mixed moderator of polyethylene particles plus liquid hydrogen, which is the first test of a mixed type moderator. The performance is better than the decoupled liquid-hydrogen moderator especially in the thermal neutron region.

As a cryogenic moderator for thermal neutron experiments, we measured neutronic performance of a poisoned liquid-methane moderator and compared the performance with that of a poisoned water moderator. The performance of the liquid-methane moderator is superior to the water moderator over the thermal region. Finally we compared the neutronic performances of a liquid-methane moderator to other various decoupled cryogenic moderators.

**I. Introduction**

In the future projects of MW class spallation sources, the development of efficient cryogenic moderators for high resolution scattering experiments in the cold and thermal regions is one of the important issues. In MW class spallation sources, as well known, monolithic solid methane cannot be used because of the burp and liquid methane will be very difficult to use because of the serious radiation damage although they have superior neutronic characteristics. The unique moderator material which can be used in MW class sources is liquid-hydrogen. However, a lower hydrogen number density of liquid-hydrogen causes the shortage of neutron intensity. A coupled liquid-hydrogen moderator with or without ambient temperature premoderator was proposed and gives 5 times higher intensity than a decoupled liquid-hydrogen moderator.<sup>(1) (2)</sup> On the other hand, the pulse width (in full width at half maximum, FWHM) of cold neutrons from a coupled moderator is much larger than that of a decoupled

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neutronic performance figure of merit

one; about two times for a moderator with a premoderator and much more for without. Therefore, this type of moderator may not be used for high resolution scattering experiments without a fast chopper at a distance very close to the source and/or a very long flight path. So, it is desired to develop a cold neutron moderator providing a relatively high intensity with a narrower pulse. For this purpose, a liquid-hydrogen moderator with decoupled premoderator was proposed as a high-efficiency cold moderator, and the neutronic performance was proved to be rather good.<sup>(3)</sup>

Recently, another method was proposed; a mixed moderator of liquid-hydrogen plus hydrogenous particles.<sup>(4)(5)</sup> This type of moderator could be used in a MW class spallation source if the solid particles can be circulated with the liquid to renew particles before suffering from serious radiation damage. This moderator can compensate the disadvantage of liquid-hydrogen, namely, a lower hydrogen number density. As solid particles solid methane and H<sub>2</sub>O ice were proposed.<sup>(4)(5)</sup> As the first step, we measured the intensity and pulse structure of neutrons from a mixed moderator consisting of polyethylene particles plus liquid-hydrogen, in order to understand the fundamental mechanism of neutron thermalization in a mixed moderator and also to know neutronic performance of this moderator. Polyethylene particles were chosen due to the reasons that this is a good substitution of water and easy to mix.

So far, the liquid-methane moderator is considered to be the best realistic moderator at a 100 kW source for high resolution experiments in the thermal region. However, the neutronic characteristics have not been well studied by experiments. So, we measured the neutronic performance of a liquid-methane moderator under the same target-moderator-reflector so far studied. We also measured the neutronic performance of the water moderator and compare the performance with that of the liquid-methane moderator.

We finally compare the measured neutronic performances of the liquid-methane moderator to those of various decoupled moderators; a B<sub>4</sub>C decoupled ambient temperature light water, a Cd decoupled liquid-hydrogen, a mixed moderator and a solid methane moderator. The results are also compared in terms of a figure of merit.

## **II. Mixed Moderator of Polyethylene Particles and Liquid-Hydrogen**

### **II. 1 Experiments**

We performed the experiments by using a cold neutron facility at the Hokkaido linac. Figure 1 shows the target-moderator-reflector assembly used. The moderator chamber has a viewed surface of 12 cm × 12 cm and a thickness of 5 cm. We used a graphite reflector of about 1m<sup>3</sup>. All moderators measured were decoupled from the reflector by 0.5 mm thick cadmium plates.

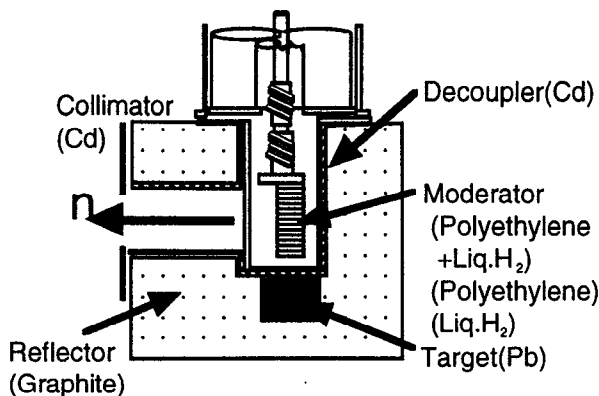


Fig. 1 Experimental setup of target-moderator-reflector assembly in study of mixed moderator.

The polyethylene particles used are low density ones with a hydrogen number density of  $7.9 \times 10^{22} \text{ H/cm}^3$ , which is almost the same as that of solid methane. The average size is about 1 mm in diam. (ranging from 0.85 to 1.18 mm). The average diameter corresponds to the mean free path in polyethylene at the Maxwellian peak energy ( $\sim 3 \text{ meV}$ ). We determined the packing factor from the

weight and the volume measurements and obtained a value of 64.5 %. The overall hydrogen number density of the mixed moderator is  $6.7 \times 10^{22} \text{ H/cm}^3$ , almost the same as that of light water ( $6.8 \times 10^{22}$ ). The average hydrogen number densities in the polyethylene particle moderator and the mixed moderator are about 1.2 times and 1.6 times higher than that of the liquid-hydrogen moderator ( $4.2 \times 10^{22} \text{ H/cm}^3$ ), respectively.

We also measured the energy spectra from two pure moderators, liquid-hydrogen and polyethylene particles for comparison

## II. 2 Results and Discussion

### II. 2 .1 Energy Spectra

Figure 2 shows the measured energy spectra. The spectrum from the polyethylene particle moderator has a peak at 6-7 meV and the intensity in the slowing-down region (approximately above 20 meV) is considerably higher than that from the liquid-hydrogen one. This is due to the fact that polyethylene has a higher hydrogen number density but no effective energy exchange mode at lower energies. The neutron intensity from the mixed moderator in the slowing-down region is slightly lower than that from the polyethylene particle moderator, in spite of a higher hydrogen number density of the mixed moderator. This can be explained as follows; in the mixed moderator neutrons are transported more efficiently to a lower energy region due to a more efficient energy transfer mechanism by mixing liquid-hydrogen. This result shows, as well known, that not only the hydrogen number density but also the energy exchange mode at lower energies play an important role to determine the spectral intensity.

The neutron spectrum from the mixed moderator is characterized by the properties of the both moderator materials, polyethylene and liquid-hydrogen. Namely, in the slowing-down region, the intensity is mainly determined by the polyethylene particles, while in the equilibrium region mainly by liquid-hydrogen.

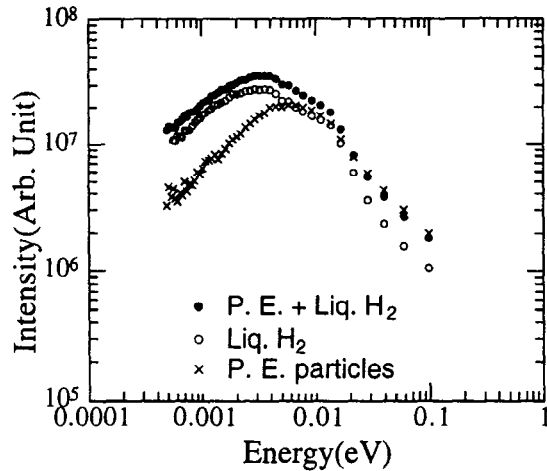


Fig. 2 Energy spectrum from the mixed moderator, compared with those from two pure moderators.

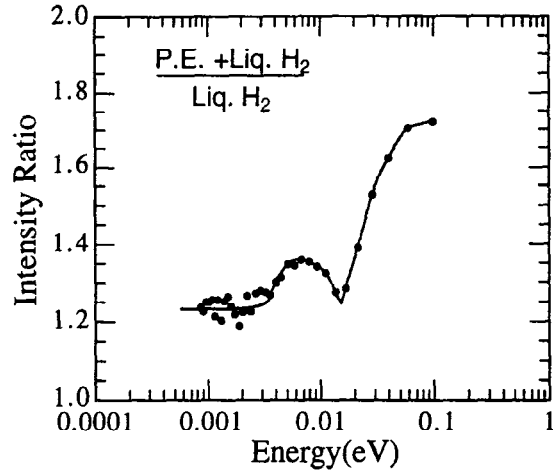


Fig. 3 Intensity ratio of mixed moderator to liquid-hydrogen moderator.

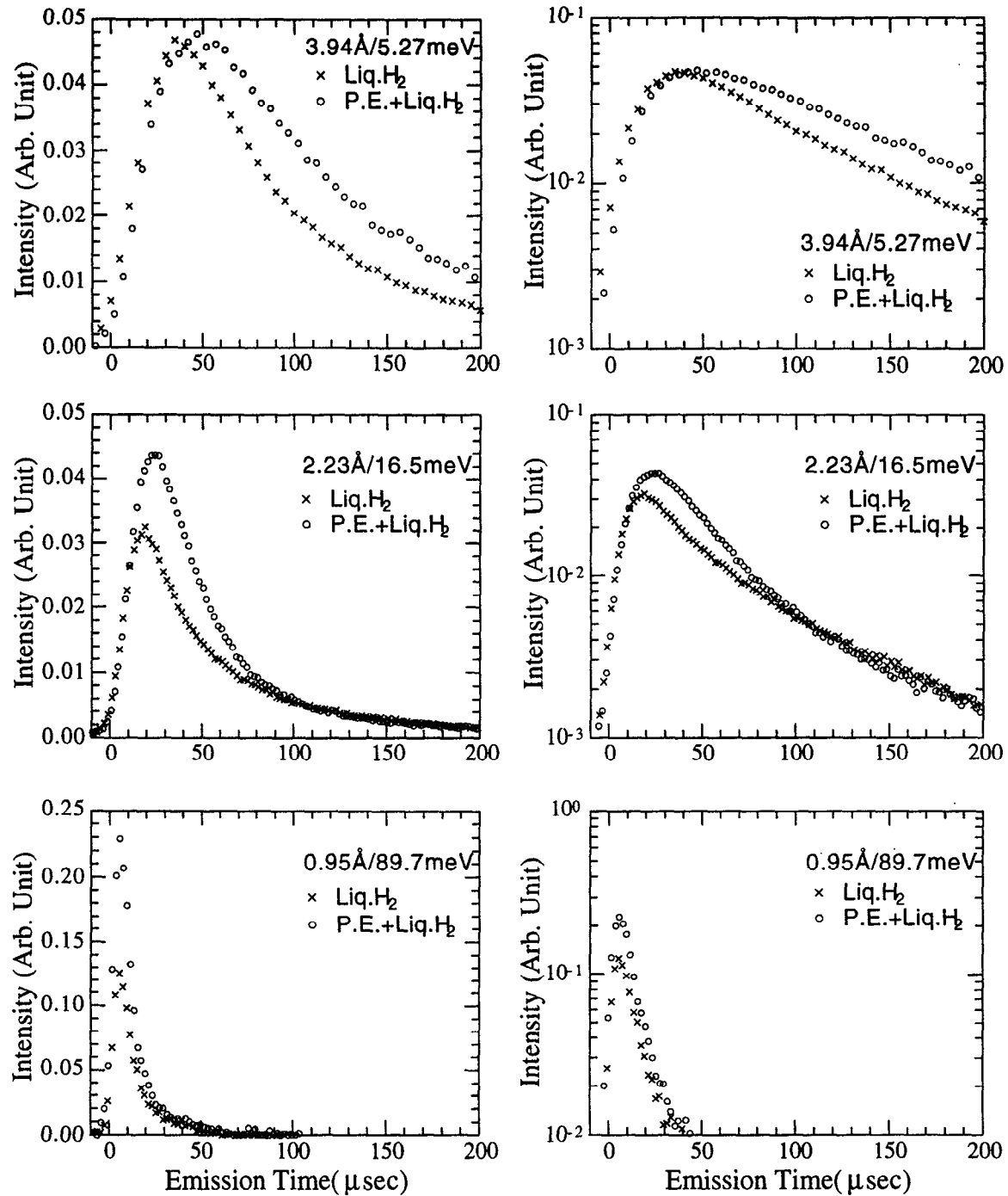
Figure 3 shows the intensity ratio of the mixed moderator to the liquid-hydrogen moderator. The neutron intensity from the mixed moderator is higher than the decoupled liquid-hydrogen moderator in all energy region. The ratio is about 1.2 at the cold neutron region but reaches to about 1.7 around 0.1 eV.

## II. 2.2 Time Distributions

Figure 4 shows measured pulse shapes from the mixed moderator and the reference liquid-hydrogen moderator at three different energies: slowing-down, transient (between slowing-down and equilibrium) and cold equilibrium regions. Those in the left side are of the linear plots, while in the right side are of the semi-logarithmic plots. In these figures, the intensities were normalized by using the time-integrated intensities shown in Fig. 2.

In pure decoupled moderators, the pulse shapes can generally be expressed by a sum of two components; slowing-down and storage terms, as reported by Ikeda and Carpenter.<sup>(6)</sup> We found that the pulse shapes from the mixed moderator can also be expressed by the Ikeda-Carpenter formula.

In the slowing-down and transient regions the pulse widths from the mixed moderator are almost comparable to those from the pure hydrogen moderator. However, the peak intensity is significantly higher. On the other hand, in the cold equilibrium region only the width increases but the peak intensity is unchanged. From these results, it turns out that in the slowing-down region the gain in the intensity is due to the increase of the pulse height but not to the increase of the pulse width. These results indicate that the characteristics of this mixed moderator is suitable for high-resolution experiments using thermal neutrons (above 20 meV) rather than cold neutrons.



**Fig. 4** Pulse shapes of mixed moderator as well as the liquid-hydrogen moderator at three energies. Left side is linear plots and right side is semi-logarithmic plots.

Figure 5 shows the pulse widths of the liquid-hydrogen and mixed moderators, in FWHM. The pulse widths of the mixed moderator is almost the same as the liquid-hydrogen one in the slowing-down and transient regions, but about 30% longer in the cold region. The pulse width broadening at lower energies is due to a higher hydrogen number density (effectively equivalent to a thicker moderator; a longer decay time) and to a poor energy exchange mechanism in polyethylene, resulting in a longer rising time as shown in Fig. 4.

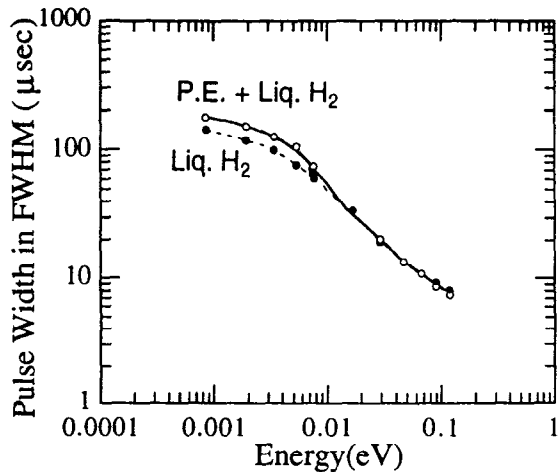


Fig. 5 Pulse width (FWHM) of neutrons from the mixed moderator as well as the liquid-hydrogen moderator as a function of neutron energy.

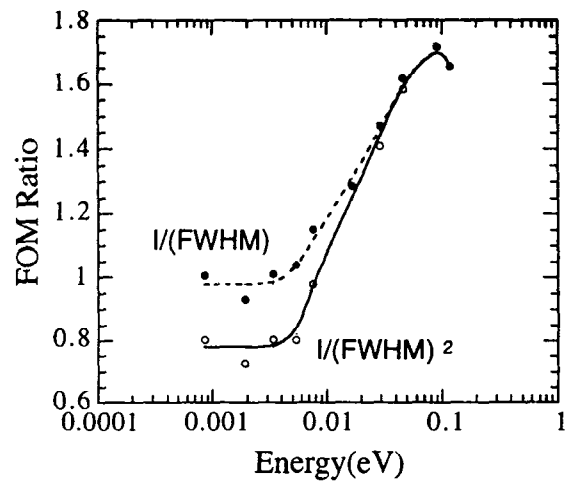


Fig. 6 Ratio of figure of merit (FOM) of the mixed moderator to the decoupled liquid-hydrogen moderator.

### II. 2. 3 Figure of Merit

We compare the measured neutronic performance to that of a decoupled liquid-hydrogen moderator in terms of a figure of merit (FOM). The figure of merits for high-resolution experiments using thermal neutrons will be defined as  $I/(\text{FWHM})^2$  since the neutron intensity incident on a scattering sample is proportional to inverse of square of flight path length (the case without neutron guide). In the cold neutron region it is difficult to evaluate the merit of a shorter flight path since in this region a neutron guide tube can be used. If we assume the merit is proportional to the inverse of the flight path length, the FOM is defined as  $I/(\text{FWHM})$ . Figure 6 shows the ratios of the FOM's, mixed moderator to the decoupled liquid-hydrogen one, calculated by both definitions. Solid line indicates the FOM value using  $\text{FOM}_1 = I/(\text{FWHM})^2$  and broken line using  $\text{FOM}_2 = I/(\text{FWHM})$ . The values will be comparable in slowing-down region, because pulse widths from the mixed moderator is almost the same as that from liquid-hydrogen moderator. The values are almost independent of the energy in the cold region, about 0.8 ( $\text{FOM}_1$ ) or 1.0 ( $\text{FOM}_2$ ), and reach at about 1.7 around 0.1 eV. This indicates that the mixed moderator is efficient rather in the thermal neutron region.

## III. Poisoned Liquid-Methane Moderator

### III. 1 Experiments

We measured the spectral intensity and pulse shapes from a poisoned liquid-methane. The target-moderator-reflector assembly used is the same as the one shown in Fig. 1 except for the moderator chamber and the decoupler. The moderator chamber has a viewed surface of 12 cm  $\times$  12 cm and a thickness of 4.5 cm. The moderator was poisoned at the center of the chamber by a 0.25 mm thick gadolinium metal plate. The moderators was decoupled from the reflector by 3 mm thick  $\text{B}_4\text{C}$  resin plates. Decoupling energy of the decoupler is about 2.5 eV. For

comparison we also measured neutronic performance of an ambient temperature  $H_2O$  moderator in the same system as the poisoned liquid methane moderator.

### III. 2 Experimental Results

#### III. 2. 1 Neutronic Performance of Liquid-Methane Moderator

Figure 7 shows measured energy spectrum from the liquid-methane moderator, compared with that from  $H_2O$  moderator. In the case of  $H_2O$  moderator, the spectra consist of  $1/E$  spectrum in slowing-down region and Maxwellian in equilibrium region. In the case of liquid-methane, there is almost flat region around 60 -100 meV.

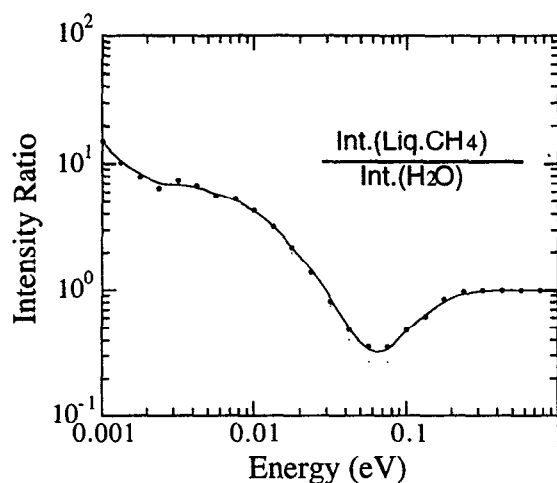
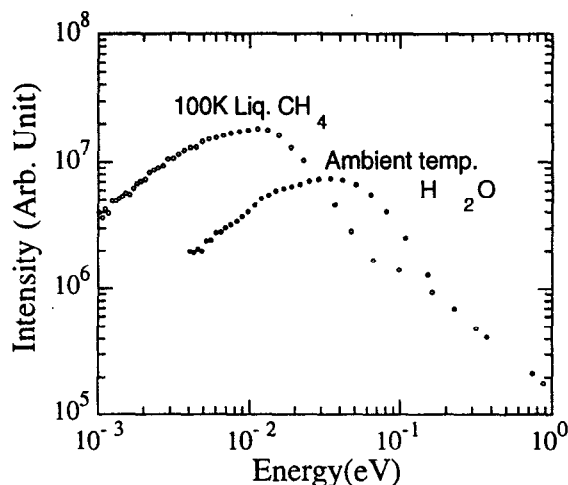


Fig. 7 Energy spectrum from liquid-methane moderator, compared with those from light water moderator.

Fig. 8 Intensity ratio of liquid-methane moderator to light water moderator.

The spectral intensity of the liquid-methane is almost the same as that from the  $H_2O$  moderator in slowing-down region due to little difference in hydrogen number density. The intensity from the  $H_2O$  moderator is higher than that from the liquid-methane moderator in the range 30-200 meV. The intensity ratio of liquid-methane moderator to  $H_2O$  moderator reaches about 10 around 1 meV, as shown in Figure 8.

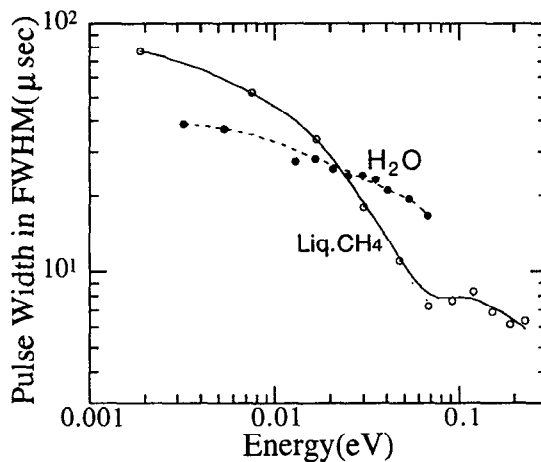


Fig. 9 Pulse width (FWHM) of neutrons from the liquid-methane moderator compared with light water moderator.

Figure 9 shows pulse width (FWHM) of neutrons from the liquid-methane moderator compared with H<sub>2</sub>O moderator. In the range 60-100 meV where the intensity from the liquid-methane is almost constant as mentioned above, the pulse width is also unchanged and in the slowing-down region above 100 meV, the pulse width would be proportional to 1/v.

The pulse width of liquid-methane is longer in the energies below 25 meV and narrower above this energy. In the energies over 70 meV, we have no data of pulse width of H<sub>2</sub>O, however, the pulse width of H<sub>2</sub>O moderator would be comparable to that of liquid-methane in the slowing-down region. The pulse width in equilibrium region is about 80 μsec in the case of liquid-methane and 40 μsec in the case of H<sub>2</sub>O.

We have calculated heat deposition in the methane-moderator in 0.6 MW case and found that the heat deposition is about 3 times higher than that in the present ISIS using Ta target.<sup>(7)</sup> If radiation damage is proportional to the heat deposition, the life will become much shorter than the present ISIS, indication that it is very difficult to use the methane moderator.

#### IV. Comparison of Neutronic Performances of Cryogenic Moderators

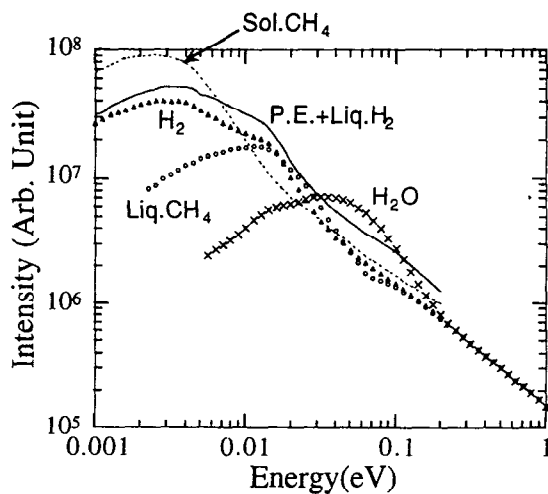


Fig. 10 Energy spectra from decoupled moderators so far measured.

A reliable comparison of the neutronic performance is indispensable for the choice of a cryogenic moderator for high-resolution thermal-neutron scattering experiments. We considered that the cryogenic moderator for high resolution experiments has to cover the energy range less than about 0.3 eV and above this energy a water moderator can provide the same neutronic performance as methane one. Our calculation indicates that we can obtain an intensity gain about 1.5 to 1.9 by reducing the decoupling energy from 2.5 to 0.5 eV. The energy spectra from five

different moderators are compared in Fig. 10; a Cd decoupled liquid-hydrogen moderator with a thickness of 5 cm, a Cd decoupled mixed moderator with a thickness of 5 cm, a poisoned liquid-methane moderator with a thickness of 4.5 cm decoupled by B<sub>4</sub>C (decoupling energy is about 2.5 eV), a Cd decoupled solid-methane moderator with a thickness of 5 cm and a B<sub>4</sub>C decoupled poisoned water moderator with a thickness of 4.5 cm.



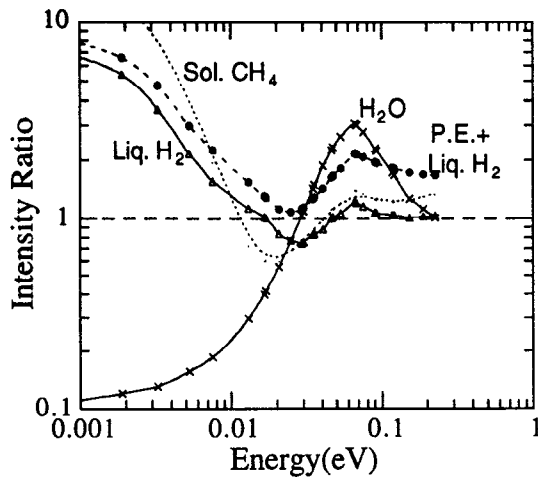


Fig. 11 Intensity ratios of various moderators to liquid-methane moderator.

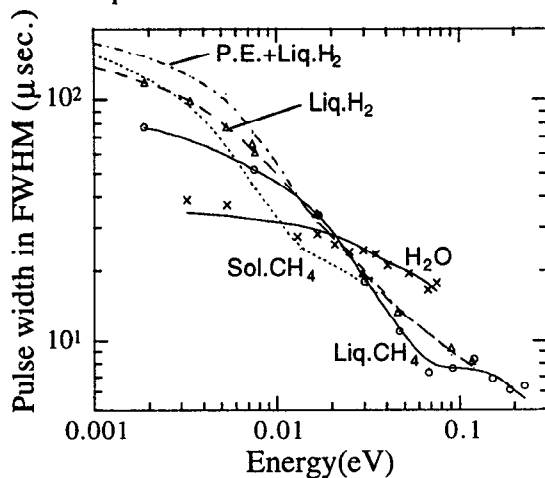


Fig. 12 Pulse Widths (FWHM) of neutrons from various decoupled moderators so far measured.

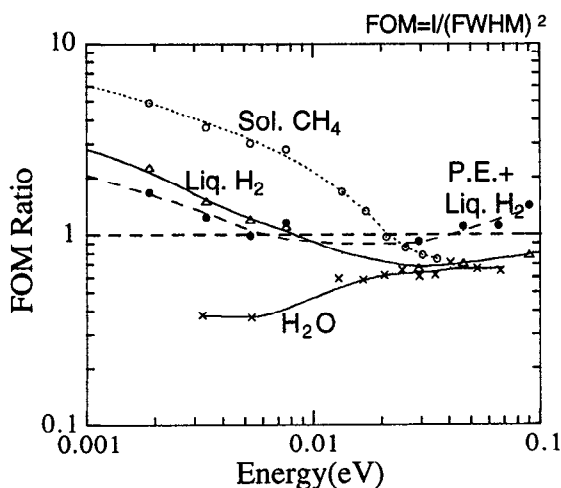


Fig. 13 Ratio of FOM's of various decoupled moderators to liquid-methane moderators.

The ratios of the intensities from moderators to that of the liquid-methane moderator are shown in Fig. 11. The pulse widths in FWHM are shown in Fig. 12. The intensity from the decoupled liquid-hydrogen is lower than that from a liquid-methane only in the region 0.015 -0.5 eV and the ratio is 0.7. Around this energy region the pulse width of the liquid-hydrogen is almost the same as or somewhat narrower than that of the liquid-methane. The intensity from the mixed moderator is higher over whole energy range. We evaluated the performance of the moderators by using the FOM defined as  $I/(FWHM)^2$  and obtained ratios to that of the liquid methane. The results are shown in Fig. 13. The FOM of the water moderator is lower than others in the energy region indicated but the FOM in the higher energy region above about 0.3 eV will be almost the same as that of the methane moderators.

The FOM of the solid methane moderator is much superior to others in the lower energy region but this cannot be used in a high power source. The relative FOM value of the liquid-hydrogen is smaller than unity above 0.01 eV but the ratio is about 0.7, which is not so bad. The relative FOM value of the mixed moderator is larger than unity below 0.005 eV and above 0.05 eV and almost the same in the mid region.

A poisoned liquid-hydrogen moderator of somewhat a larger total volume will increase the intensity without changing the pulse width. Such experiments are important.

## V. Conclusion

As a high-resolution (short pulse) moderator, a mixed moderator of liquid-hydrogen plus polyethylene particles exhibits a good neutronic performance compared with a liquid-hydrogen moderator, especially in the thermal neutron region. Therefore, this type of moderator will be suitable for high resolution experiments in the thermal region.

A liquid- methane moderator has good neutronic characteristics but the superiority is not so large compared with the liquid- hydrogen moderator. Furthermore, it is very difficult to use a liquid-methane moderator in a MW class spallation neutron source since the radiation damage in the methane moderator is too large.

A poisoned liquid-hydrogen moderator will be one of the most promising candidates as moderator for high resolution thermal neutron experiments. This type of moderator will be studied near future. For the long term study the development of an advanced type cryogenic moderators for example, a mixed moderator, is important.

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