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NEUTRONIC STUDIES OF A LIQUID HYDROGEN-WATER COMPOSITE MODERATOR

T.Tahara*, E.B.Iverson, J.A.Crabtree A,T.Lucas, M.Ooi*, H.Iwasa*, and Y.Kiyanagi*

Oak Ridge National Laboratory, Oak Ridge, TN37831
*Faculty of Eng., Hokkaido University, Kita-13 Nishi-8, Kita-ku, Sapporo 060, Japan

Abstract

A liquid hydrogen-liquid water composite moderator may provide performance like liquid methane at high-power spallation sources where liquid methane is impractical. We have measured the neutronic properties of such a composite moderator, where a hydrogen layer 1.25cm thick was closely backed by water layers of 1.75cm and 3.75cm thickness. We also studied a moderator in which a 1.75cm water layer was closely backed by a 1.25cm hydrogen layer. We further performed simulations for each of these systems for comparison to the experimental results. We observed enhancement of the spectal intensity in the "thermal" energy range as compared to the spectrum from a conventional liquid hydrogen moderator. This enhancement grew more significant as the water thickness increased, although the pulse shapes became wider as well.

1. Introduction

High-power spallation neutron sources, such as the Spallation Neutron Source Project to be constructed at Oak Ridge National Laboratory, have high radiation damage rates in most of the neutronically significant components, including the moderators. This is especially important for moderators composed of liquid methane. In the liquid state (around 100 K) methane suffers from severe problems with radiation-induced polymerization, to such a level that the highest-power existing spallation sources must significantly limit the lifetime of their liquid methane moderator vessels, imposing significant constraints on operational strategies for the facility. At damage rates corresponding to the Spallation Neutron Source (2~MW of proton power on a mercury target) the use of liquid methane does not currently appear to be practical, even though it provides performance characteristics generally agreed to be highly desirable. In fact, most "new" spallation source projects, when they develop proposed instrument suites, assign approximately half of the initial instruments to liquid methane moderators when that choice is offered. As no practical means to use liquid methane at these damage rates has been demonstrated, we have explored an alternative moderator concept, the hydrogen-water composite moderator, as an alternative to liquid methane, one which would be tolerant of radiation.

As part our study of a composite moderator, we have constructed a prototype moderator and measured its performance at the University of Hokkaido Electron Linac facility. These prototype experiments are essential to realistically judge the composite moderator usefulness, since many aspects of the composite moderator performance depend on how closely the hydrogen and water layers can reliably be placed while still maintaining their desired temperatures and states.

2.General Description of Composite Moderator

The experiments were performed at the 45 MeV Electron Linac facility at the University of Hokkaido. An outline of the experimental system is shown in Figure 1. The target is lead, and is 7.5cm by 8cm by 15cm. A graphite reflector of thickness at least 40 cm surrounded the target and moderator. A decoupler layer of B4C 3mm thick (corresponding to a decoupling energy of 2.5eV) surrounded the moderator. A schematic diagram of the moderator vessel, showing major dimensions of the various regions, appears in Figure 2. There are 2 chambers which can be filled with water on each side of the liquid hydrogen chamber. We continuously circulated water through the filled water chambers (different chambers were filled during different parts of the experiment) both to keep the temperature constant and to prevent the water from freezing. The water chambers could be filled with water and emptied of water remotely in order to select the thickness of water on either side of the hydrogen vessel from the options of 0cm (no water on a given side), 1.75cm, and 3.75cm. The water temperature in all cases is the ambient room temperature, about 295K. The temperature of the hydrogen remained at about 18K for all experiments.

We performed experiments on the five moderator configurations indicated in Figure 3. The five cases are:

- o H_2 with 3.75cm H_2O backing (No.1),
- o H_2 with 1.75cm H_2O backing (No.2),
- o 3.75cm H_2O with H_2 backing (No.3),
- o 1.75cm H_2O with H_2 backing (No.4) and
- o H_2 alone(No.5).

We performed Measurements of spectral intensities and pulse shapes for all arrangements except for No.3, since it was expected that the pulse shape for this configuration would be almost the same as a water moderator, and experimental time was somewhat short.

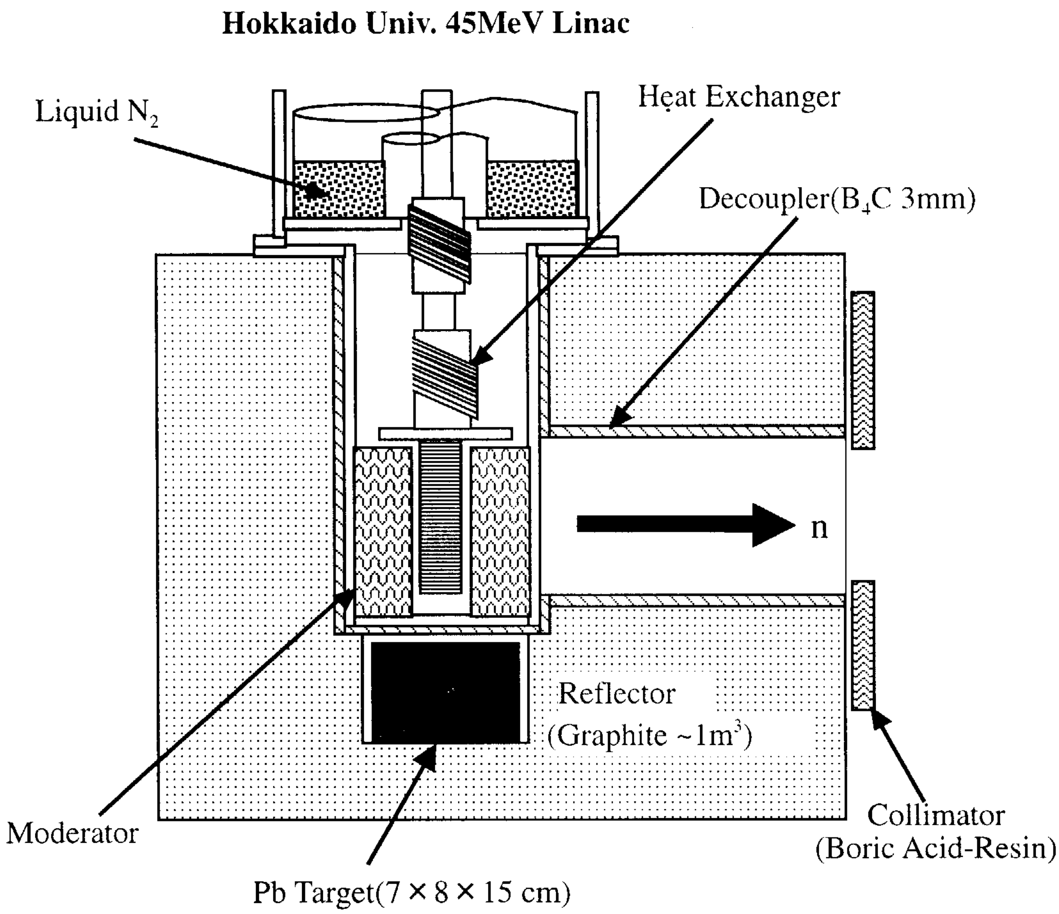

Fig.1 Experimental setup

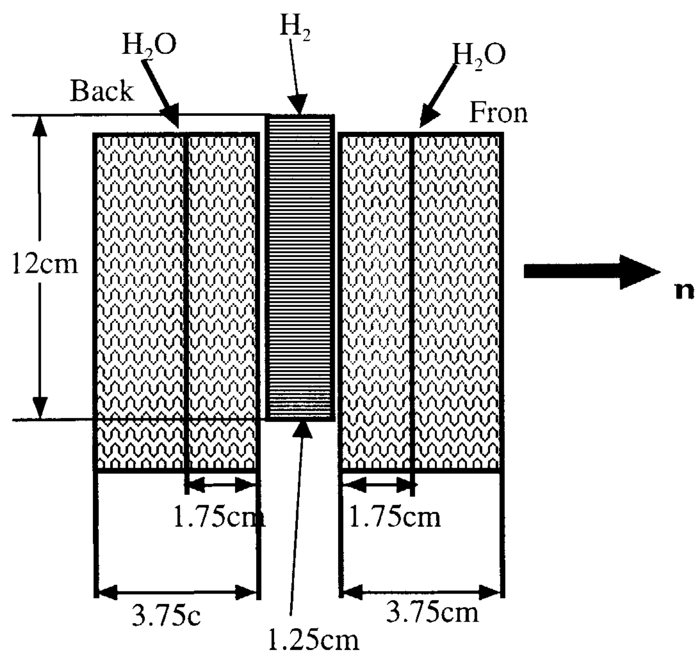

Fig.2 Moderator chamber assembly

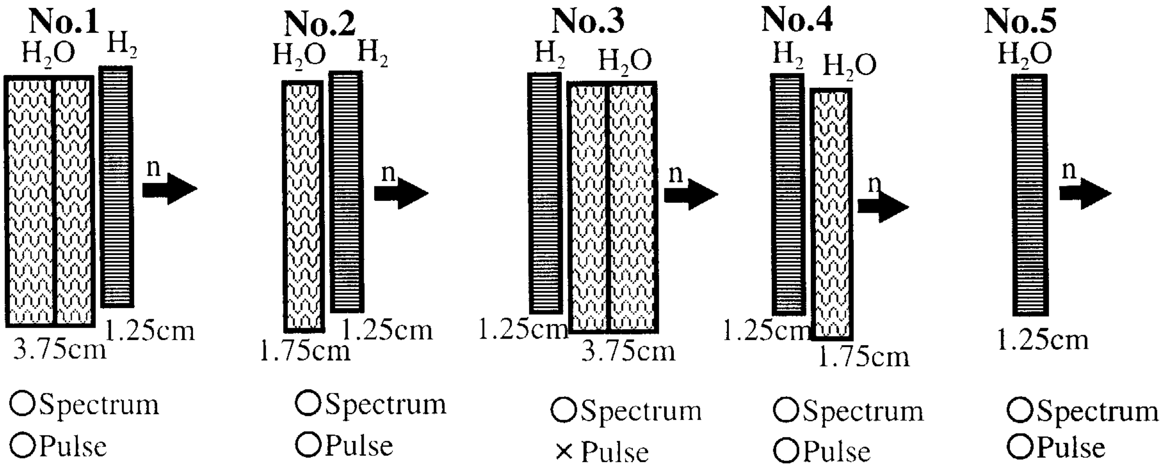

Fig.3 Moderator systems used in experiment

3.Experimental Result

The measured energy spectra are shown in Figure 4. When the neutron beam is extracted from the hydrogen side of the moderator we can clearly see a large increase in neutron intensity for the composite moderator, when compared with the hydrogen alone, over the energy regions observed. This increase is thus due to the water layer placed behind the hydrogen (configurations 1 and 2 in Figure 3). Furthermore, the composite with the thicker water layer gives about 1.8 times the intensity below 90 meV than the composite with the thinner water layer, as shown in Figure 5.

As expected, the spectrum of the ambient temperature water is dominant in the systems (numbers 3 and 4 in Figure 3) where the composite was viewed from the water side. Figure 6 shows pulse shapes from three moderators; H_2 alone, H2 with 1.75cm water backing and H_2 with 3.75cm water backing, at three different energies: 7.3 meV, 30 meV, and 90 meV. The pulse rises more quickly for a single H_2 moderator than for composite moderators, so the peaks of the H_2 moderator pulses appear at earlier times than do the composite ones. Decay rates are much slower for the composite moderator than for hydrogen, but the peak intensities of the composite moderator are higher. This moderator does appear to work as a dual characteristic moderator; namely, a thermal moderator when viewing the water side and a kind of cold moderator when viewing the hydrogen side.

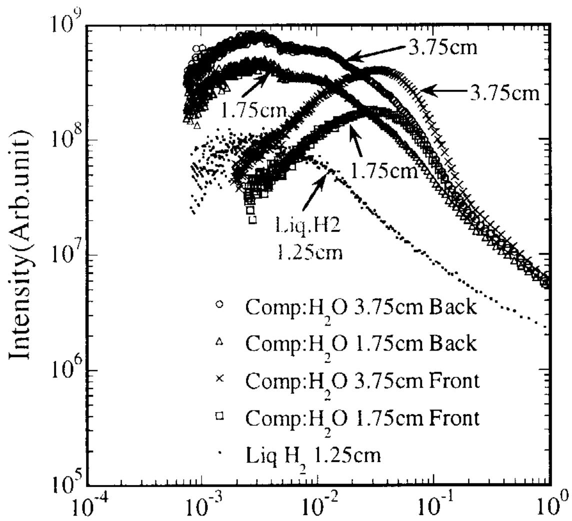

Fig.4 Observed spectra from the composite moderators

4.Comparison of Calculational and Experimental Results

We performed simulations of the composite moderator experimental setup using HMCNP4B with ENDF/B-V and VI cross section data. Our calculations used a simplified geometry as shown in Figure 7, simulating our experimental setup. First, we compared the spectral shapes and relative intensities. To compare relative intensities, we normalized the calculated spectra of the liq.H_2 with 3.75cm H_2O backing so as to give the same value as the experimental measurement at 1eV, as shown in Figure 8.

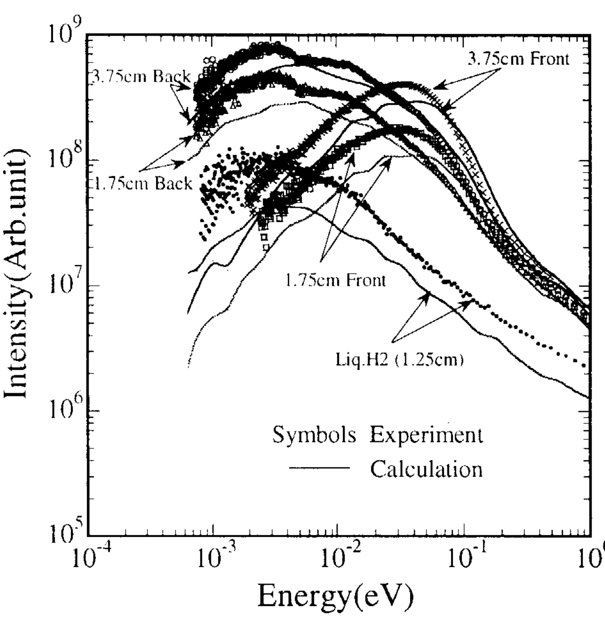

Fig.8 Comparison of energy spectra obtained by experiments and calculations

There are some differences between the measured and calculated spectral shapes, not only for the spectra observed from H_2 side, but also for those observed from H_2O side. In general, the energy spectrum falls of with decreasing energy faster in the calculated spectra than in the measured spectra. The measured spectral peak locations (proportional to the spectral temperatures) are lower in the experimental values. In all cases, the calculated spectral intensities at low energies are lower than the experimentally measured intensities. We observe a big difference in the calculated and measured spectra in the case of the 1.25cm thick hydrogen moderator.

The reason for this discrepancy is still unclear. It is likely partly due to over-simplification of the target-reflector-moderator assembly geometry in the simulation. There may have been some water remaining in supposedly empty water chambers, which might especially effect the intensity of the thin hydrogen moderator. We are still trying to clear up this discrepancy.

We also compare pulse shapes, measured and calculated, in Figure 9, where they are normalized to the same peak intensity, in order to see differences in shape. Overall, the shapes are similar. The time-decay of the calculated pulse shapes is faster than that in the experiments. We compare pulse width (full width at half maximum) in Figure 10. The calculated results almost agree with experimental ones at higher energy but discrepancies appear in the lower energy region.

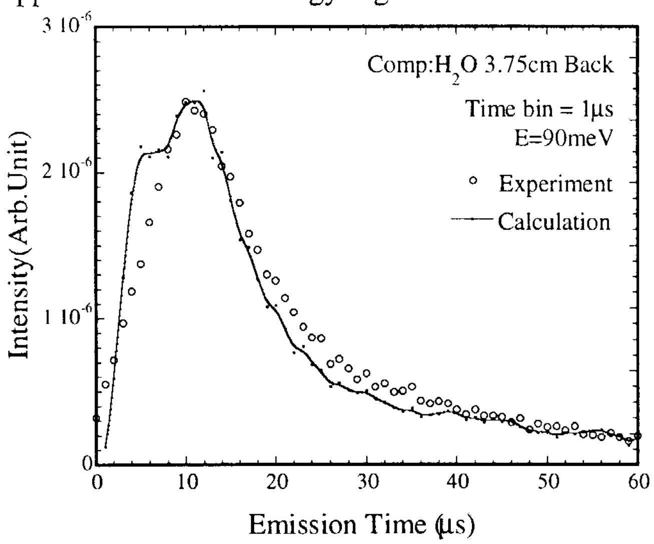

Fig.11 Double peak appeared at high energy

We observe a "double peak" shape in calculated pulse shapes at higher energies (above 80--90 meV), perhaps since neutrons emitted from the water layer can be directly observed through the hydrogen layer. This might cause a difference in flight time across the distance between the viewed hydrogen surface and the viewed water surface. Such a calculated result is shown in Figure 11 (black points with line), which indicates a shoulder on the rising edge of the pulse shape. We performed one set of pulse shape measurements in a higher resolution mode, with a 1μs electron pulse width and a 1μs channel width on the time analyzer, in the hopes of seeing any indication of this double peak behavior. We could not observe any double pulse peak similar to the calculated one, although we did observe the more gently rising edge in the composite moderator pulse shape than in that of the hydrogen moderator. This is probably due to the fact that the hump in the rising side of the pulse shape is smeared out by the relatively wide experimental time widths and instrumental resolution.

5.Comparison of Experimental Results to Liquid Hydrogen 4.5cm Moderator

We compare in Figure 12 the spectral intensity of the composite moderator with that of a 4.5cm thick hydrogen moderator. The intensity of the composite moderator with 3.75cm H_2O layer is higher in the thermal energy region due to the contribution of the thermal spectrum of the water moderator behind the H_2 moderator. The intensity ratio of these two moderators is shown in Figure 13. We clearly see that the composite moderator gives higher intensity only in the thermal energy region. The largest intensity gain is about a factor of 2. We compare pulse shapes in Figure 14. The pulse shapes from the composite moderator rise and decay slowly when compared to the hydrogen moderator. At low energy, the pulse shape of the composite moderator is similar to that of 4.5cm thick H_2 moderator, although the peak intensity of the composite moderator is much less than that of the H_2 moderator. The FWHM of the composite moderator pulse shapes at lower energies are much wider than those of the 4.5cm H_2 moderator.

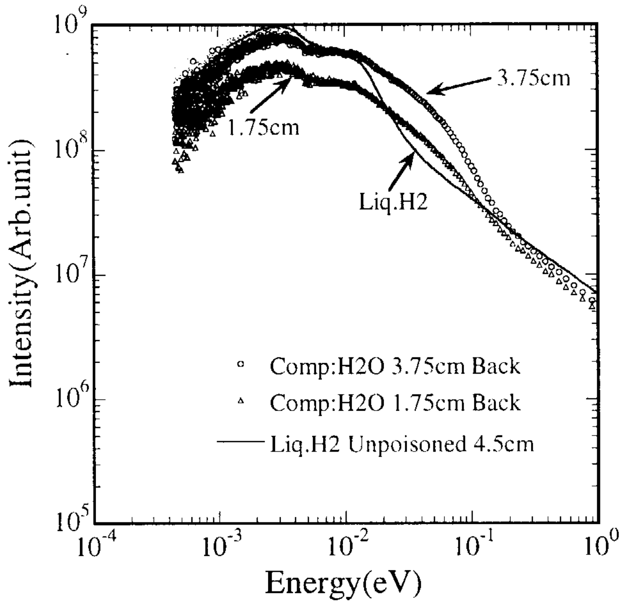

Fig.12 Comparison with a 4.5cm H_2 moderator

6.Effect on Scattering Instrument Designs

Of course, the most important aspect of moderator performance is how well that moderator satisfies the needs of scattering instrument designs. The reason we chose to look at the composite moderator in the first place was as a possible replacement for liquid methane, which is usually used for relatively high resolution scattering instruments. Thus the wider pulses observed from all composite moderator designs, both in calculation and experiment, are a serious disadvantage. This disadvantage probably outweighs most advantages gained from the improved spectral intensity of a composite moderator. Even if the double pulse shape seen in calculations is never present in experiment, as it was not in our experiments here, the resolution effect alone might be an problem which cannot be overcome. The composite moderator does, however, always have intensity superior or equal to a hydrogen moderator, depending on energy, and has significantly higher intensity at low energies than any water moderator. Thus, if resolution were not an issue for a particular instrument design, and that instrument desired high intensity at all energies, it might very well benefit significantly from the composite moderator.

7.Conclusion

We have measured the neutronic performance of a variety of hydrogen-water composite moderator configurations, and compare these measurements to the results of simulations. We find that the simulation techniques do a reasonable job of predicting composite moderator performance, but that there are significant discrepancies between simulation and measurement.

We do not measure any "double peak" pulse shapes for any composite moderator configuration in the energy range studied (below some 100 meV), in contrast to simulations performed as part of this study and elsewhere. We do measure relatively broad pulse shapes for our composite moderator configurations, broader than typical water or hydrogen pulse shapes. These broader pulse shapes indicate that the composite moderator may not be able to replace liquid methane for high resolution neutron scattering instruments. However, instrument concepts which call for high intensity at all energies and are not limited in resolution by the moderator pulse shape might benefit from the composite moderator.

Acknowledgement

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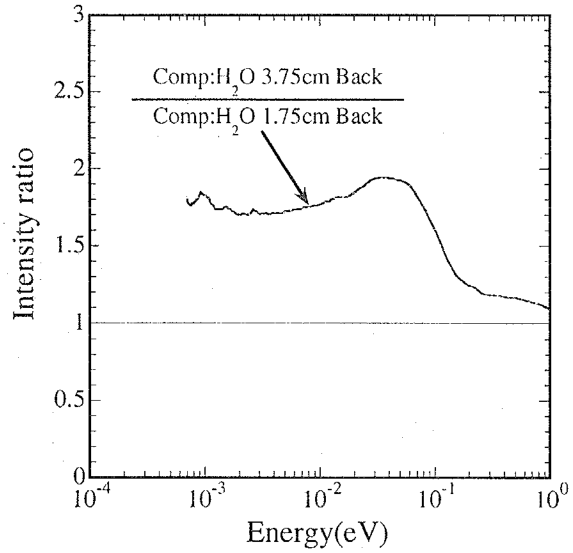


Fig.5 Intensity comparison between composite moderators with 3.75cm and 1.75cm water

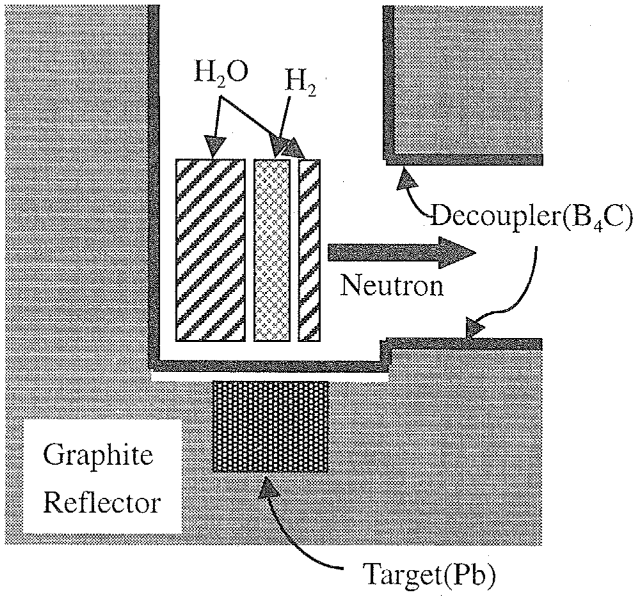
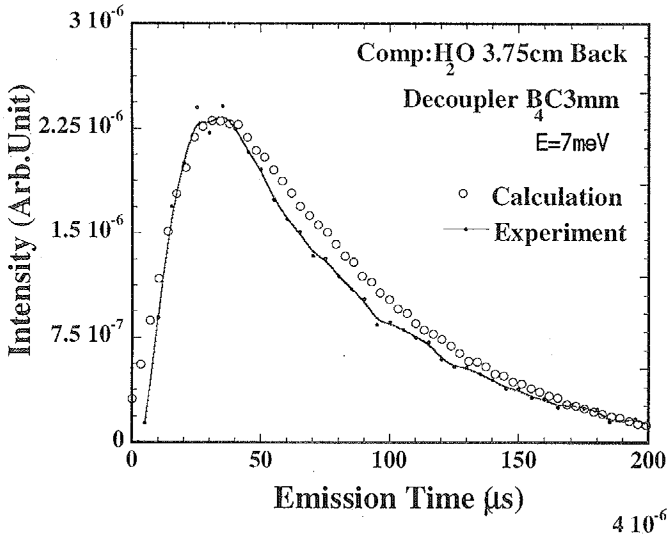


Fig.7 Simulation Geometry



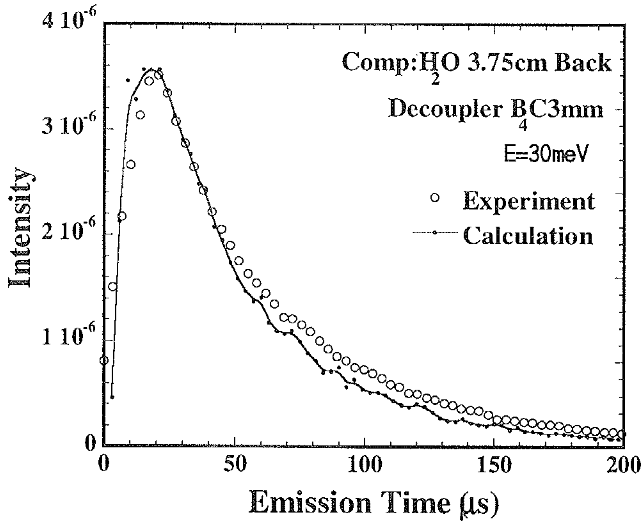


Fig.9 Comparison of pulse shapes obtained by experiments and calculation

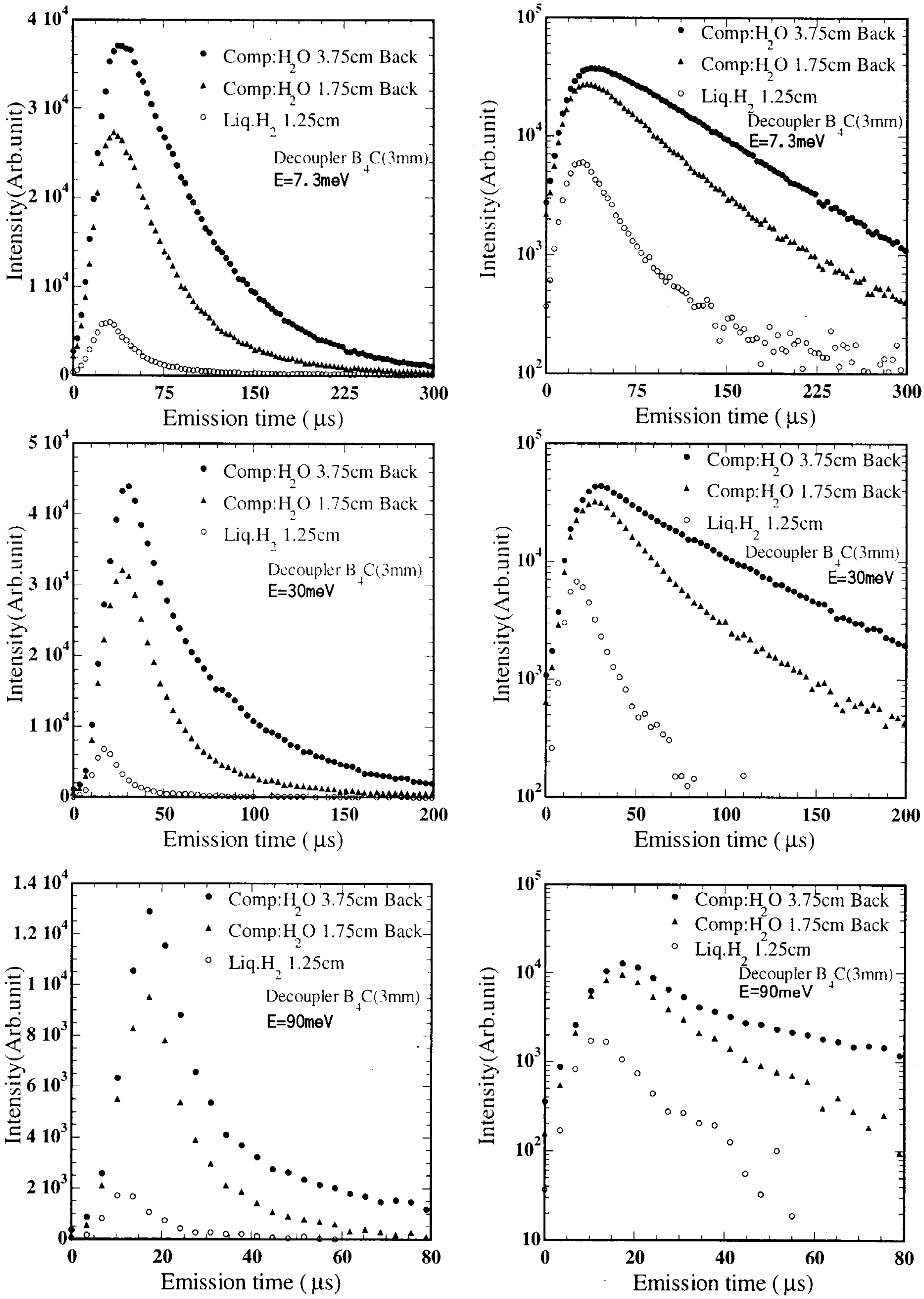


Fig.6 Comparison of the pulse shape from composite and liq,H_2(1.25cm) moderators
Left side is linear and right semilogarithmic (Experiments)

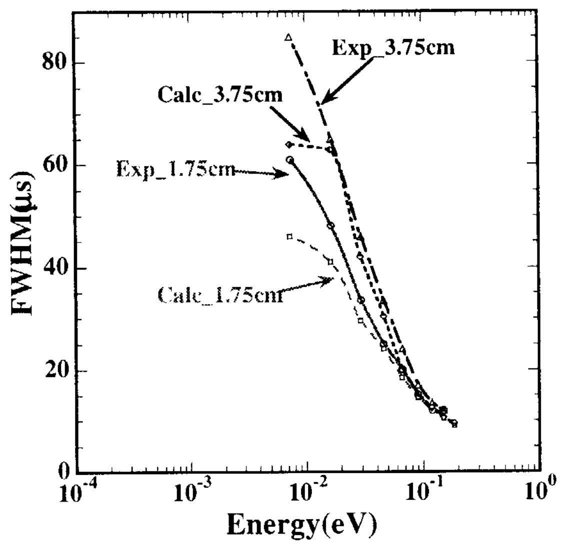


Fig.10 Comparison of FWHM

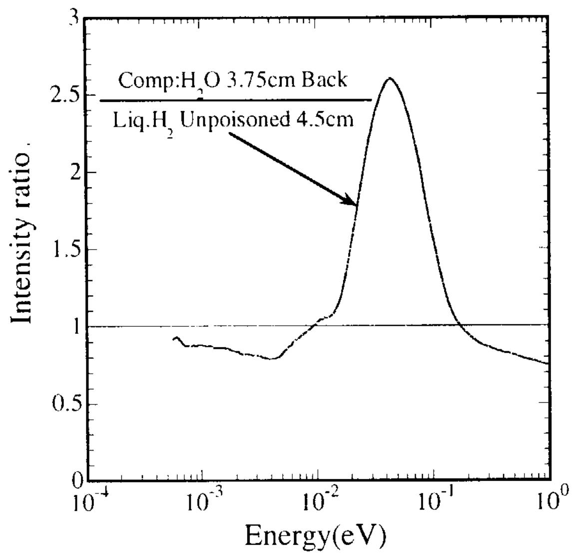


Fig.13 Comparison of intensity between the composite and a 4.5cm H_2 moderators

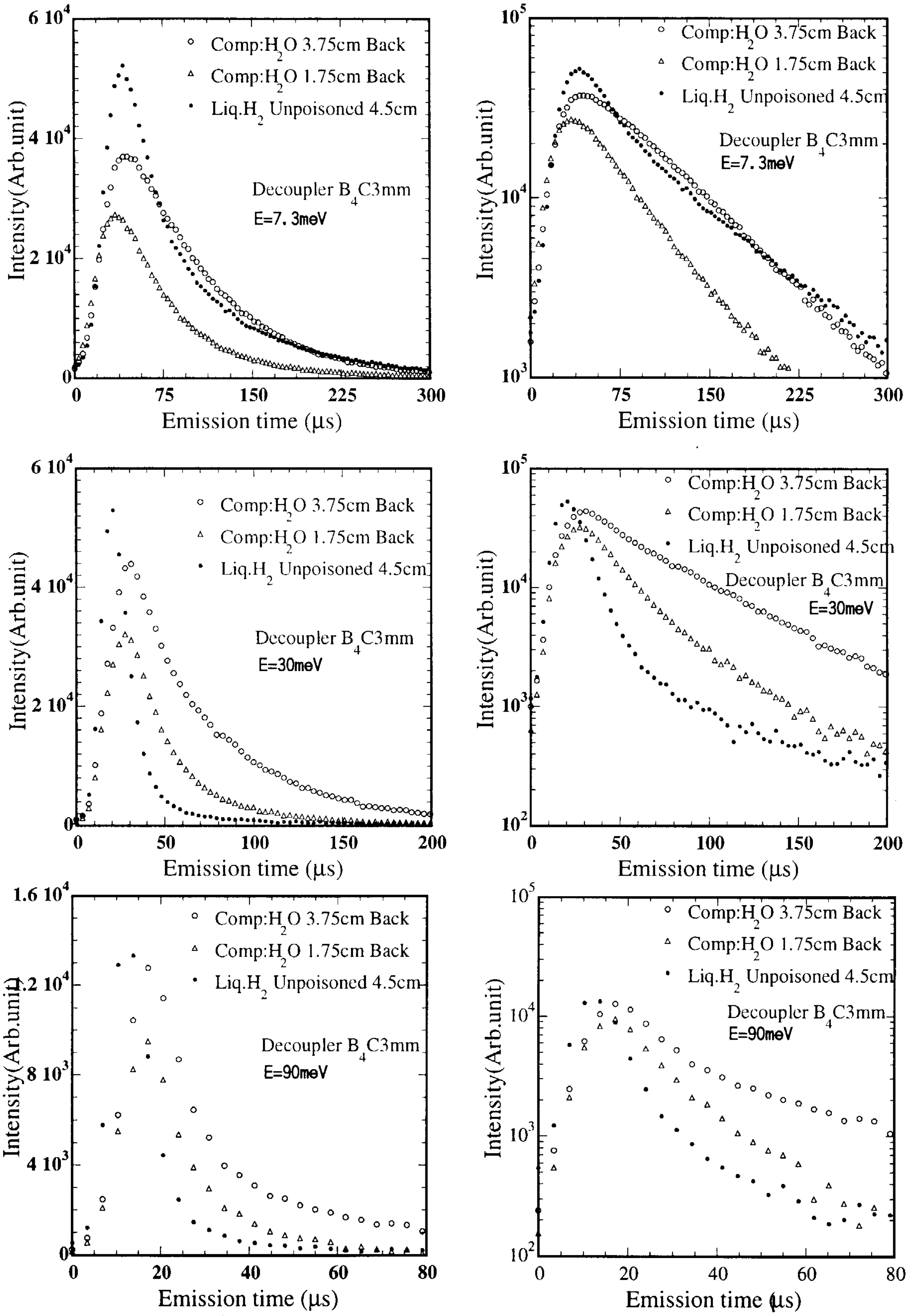


Fig.14 Comparison of the pulse shape from composite and liq.H_2 (4.5cm) moderator