Studies of Decoupled Composite Moderators of Liquid Hydrogen and Zirconium Hydride

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Abstract

We report the results of a series of measurements of the intensities and pulse shapes of decoupled composite 2 cm-thick liquid Hydrogen moderators, with various thickness of cold Zirconium hydride backing.

Introduction

The purpose of these studies is to explore the possible alternatives to liquid methane as a moderator to provide short pulses of thermal and cold neutrons in high power pulsed spallation neutron sources. The attractive properties of liquid methane in this application are well known, which follow from its reasonably low temperature in the liquid state ($T_{melt} \sim 90°K$), its high proton density and its excellent thermalizing power extending to small energy transfers ($\Delta E_{rot} \sim 1.$ meV). However, radiation damage in liquid methane leads to the release of Hydrogen gas and to the accumulation of solids which obstruct flow and diminish heat transfer. These problems are observable but manageable in present-day facilities, but especially the latter gravitates against the use of liquid methane in the high power facilities that are now being designed.

Liquid Hydrogen has the advantage that it is insensitive to radiation damage, in that the products of radiolysis rapidly recombine into the parent material (although the low-temperature para form tends to revert to the high-temperature normal ortho-para mixture under intense irradiation). Additionally, Hydrogen remains liquid to lower temperatures ($T_{melt} \sim 15°K$) than methane and exists at near liquid density as supercritical vapor at reasonable pressure ($p_{crit} \sim 15$ atm) still at low temperature ($T_{crit} \sim 35°K$), which are engineering advantages. The disadvantages of liquid Hydrogen are also well known, namely its relatively low proton density and its relatively poor thermalizing power ($\Delta E_{rot} \sim 15.$ meV), which inhibits thermalization to the low temperature of the liquid.

Zirconium hydride, ZrH_2, on the other hand, has a high proton density and is also stable at high levels of irradiation and at reasonably high temperatures; engineering data exist in relation to its use in TRIGA reactor fuel. However, ZrH_2 lacks thermalizing power at low energies because the proton is so tightly bound in the metal lattice ($\Delta E_{vib} \sim 130.$ meV). Composite moderators in which the viewed layer is of liquid Hydrogen and the backing is of Zirconium hydride have the prospect of providing the high efficiency and short epithermal pulse structure that follow from the high proton density of the backing material, and the reasonably low spectral temperature and efficient thermalization that characterize liquid Hydrogen. Furthermore, the cryogenic material in a composite moderator will be of smaller volume than in a monolithic moderator, so that the cryogenic cooling requirements are reduced in proportion to the volume, since the backing material can assumedly be cooled separately and less aggressively. Still further, depending on the relative positions of primary neutron source, moderator and backing, the backing may function as premoderator, further reducing the cryogenic heat load. The present work extends the earlier investigations of composite 5 cm thick liquid Hydrogen moderator systems with ambient temperature ZrH_2, H_2O and polyethylene premoderators.

We have investigated systems consisting of liquid Hydrogen 2 cm thick with 0 to 3 cm thick Zirconium hydride backings, reflected with graphite and decoupled with Cadmium. We have measured spectral intensities for wavelengths between 0.3 and 13. Å, and determined the pulse shapes for neutrons of wavelength from 1. to 10 Å. We present pulse widths and comparably-normalized spectra as functions of wavelength for the studied systems, and provide detailed pulse shapes for selected wavelengths.

Experimental Arrangements

The Hokkaido University 45 MeV electron linac provided a pulsed fast neutron source for the measurements, operating at approximately 50 Hz, with 0.3 μA time average current and 20 nsec pulse width for energy spectrum measurements and at 30 μA current and 3 μsec pulse width for pulse shape measurements. The target was a water-cooled lead block. Figure 1 shows the moderator-reflector assembly, which consisted of a graphite reflector approximately 1 × 1 × 1 m^3 in volume, a 2 × 12 × 12 cm^3 18 K liquid Hydrogen moderator. Packed Zirconium hydride powder, at a hydrogen number density of 0.64×10^{24} gm/cm^3, was contained in Aluminum cans. A thermocouple in the backing moderator registered the temperature, which was the same as that of the Hydrogen in these studies. Cadmium apertures defined the 10 × 10 cm^2 viewed area of the moderator, and the beam path void was lined with Cadmium. The electron beam struck the target in the direction perpendicular to the plane of the figure.

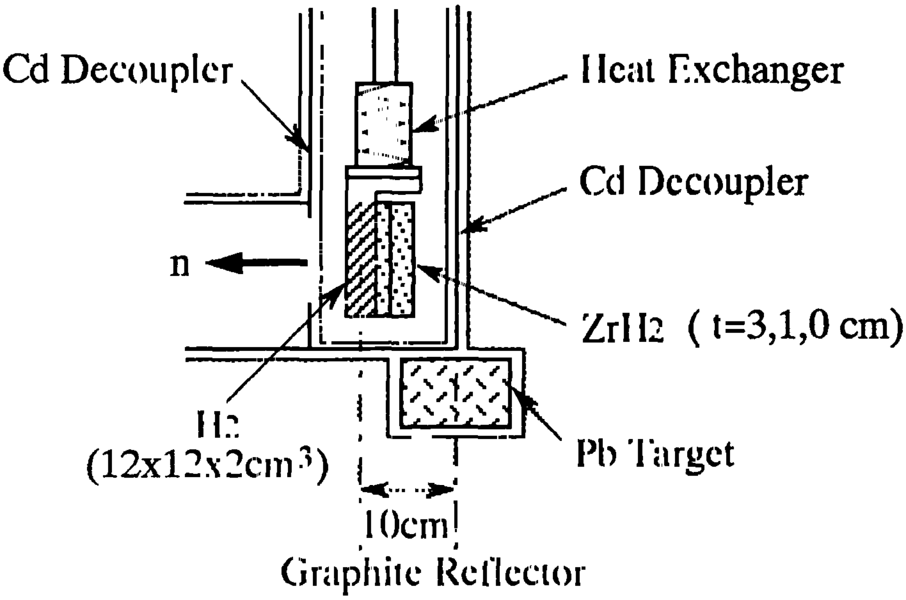

Fig. 1 Decoupled composite Liquid Hydrogen -Zirconium hydride moderator system

The measuring system for the spectra and pulse widths was the same for these measurements as is described elsewhere(1). The time resolution of the analyzer system was approximately 0.1 %. Time channels were 5 μsec long for the pulse shape measurements.

Energy Spectra

Figure 2 shows the flux per unit energy normalized to the same electron beam current and for fixed target and liquid Hydrogen positions. Also shown is the spectrum measured earlier for a 5-cm-thick liquid Hydrogen moderator, approximately normalized for the same conditions. At low energies the shapes of the spectra are almost identical. The intensities increase with increasing thickness of ZrH_2; for 3 cm thickness, the intensity is almost the same as for 5 cm thick liquid Hydrogen with no premoderator.

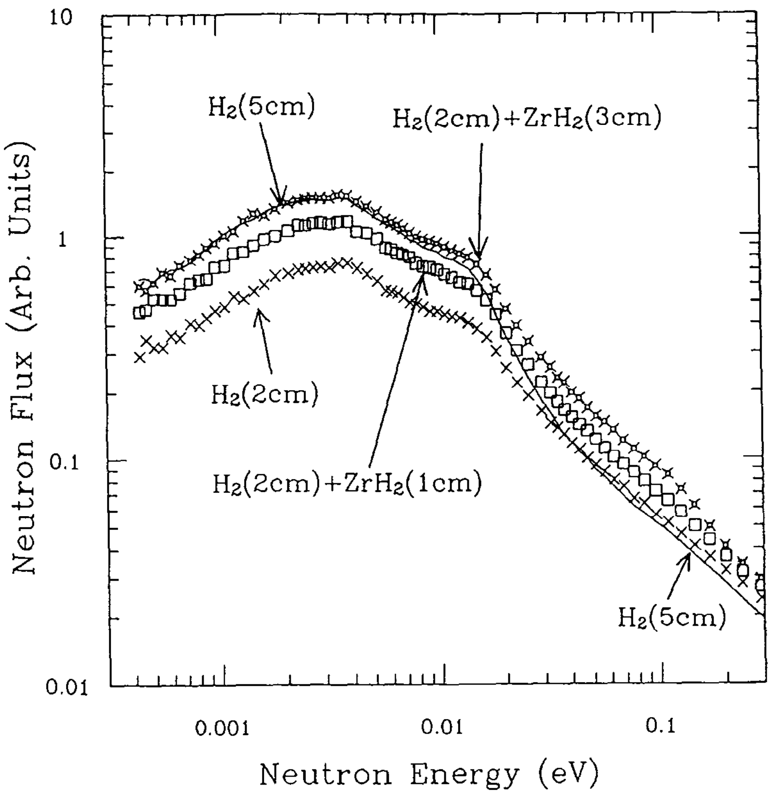

Fig. 2 Flux per unit energy for 2 cm thick liquid Hydrogen moderators with 0, 1 and 3 cm thick Zirconium hydride premoderators, and for a 5 cm thick liquid Hydrogen moderator with no premoderator.

Figure 3 shows the ratios of the spectral intensities for the cases with ZrH_2 to the intensity for the case for 2 cm L-H_2 only. The intensity gain factor at low energy is a factor of 2 for the 3 cm thick ZrH_2 case.

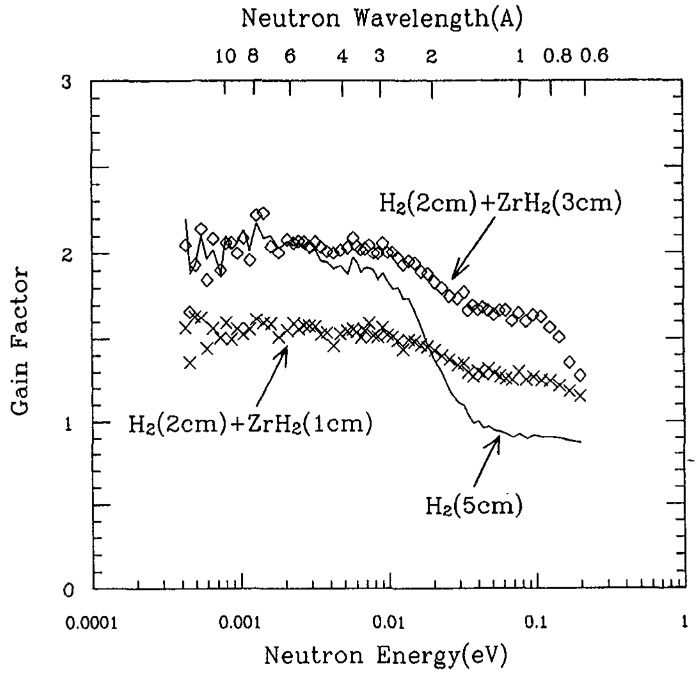

Fig. 3 Ratios of fluxes of liquid Hydrogen moderators with Zirconium hydride premoderators of various thickness and of a 5 cm thick liquid Hydrogen moderator with no premoderator, to the flux for the 2 cm thick moderator with no premoderator.

Pulse Shapes

Figure 4 shows the pulse shapes for three representative wavelengths, in linear and similogarithmic form, from the three different moderator systems and for the 5 cm thick liquid Hydrogen moderator. The time scales have been adjusted to correspond to the emission time, that is, the time that neutrons crossed the viewed surface of the liquid Hydrogen, by subtracting the known time of flight across the path from the moderator to the detector. The intensities for the three studied moderators are consistently normalized.

For each wavelength, the widths of the pulses and the peak intensities increase slightly as the thickness of the ZrH_2 layer increases. The changing width is almost entirely due to the change in the length of the decay time. The shapes of the leading edges are almost identical; we believe that this represents the real situation, because the rise times are longer than the resolution time of the analyzer system.

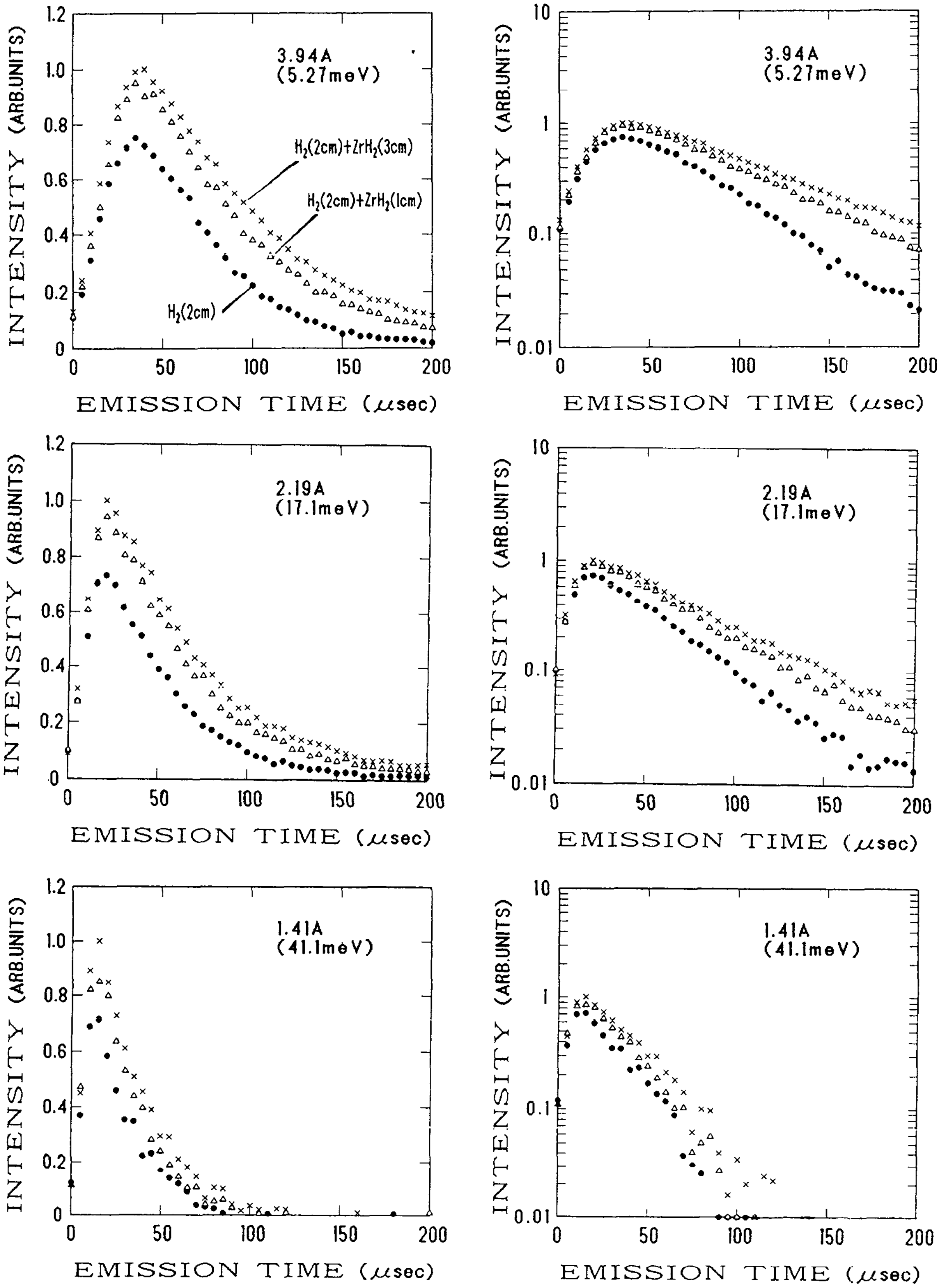

Fig. 4 Pulse shapes for neutron of 3.94, 2.19 and 1.41 Å wavelength, for 2 cm thick liquid Hydrogen moderators with 0, 1 and 3 cm thick Zirconium hydride premoderators.

Figure 5 shows the pulse FWHM as a function of wavelength, determined from data on all the wavelengths measured. For comparison, the figure also shows FWHM for the 5 cm L-H_2 and for 5 cm thick L-CH_4(2) and poisoned L-CH_4(3)(4)moderators. The widths of the pulses of composite moderators studied are less than the widths for the 5 cm L-H_2 case at longer wavelengths.

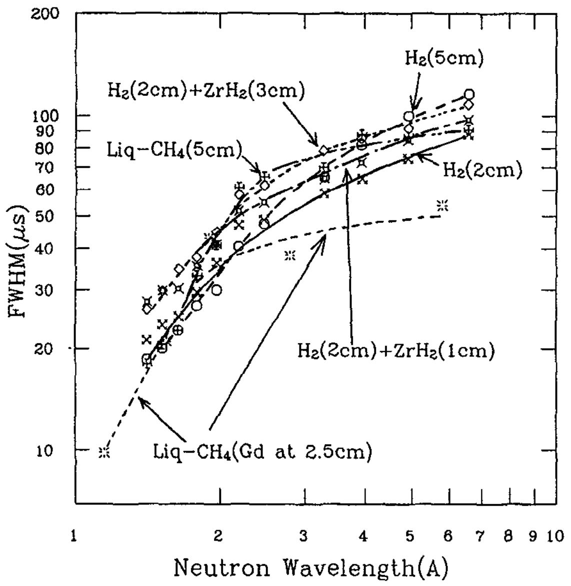

Fig. 5 Pulse FWHM as a function of wavelength for neutrons from 2 cm thick liquid Hydrogen moderators with 0, 1 and 3 cm thick Zirconium hydride premoderators, and from a 5 cm thick liquid Hydrogen moderator with no premoderator. For comparison, the figure also shows the width of pulses from a 12 × 12 × 5 cm liquid methane moderator and a 10 × 10 × 5 cm liquid methane moderator, poisoned with Gd 2.5 cm below the surface.

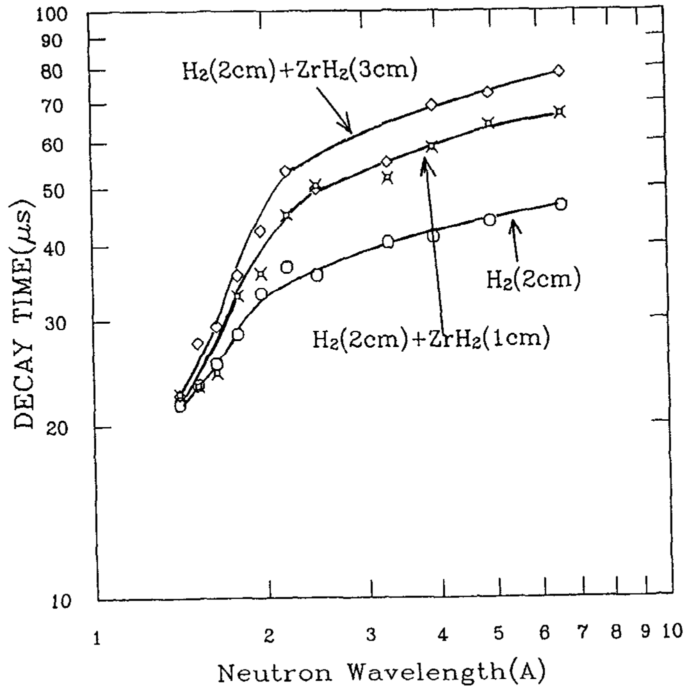

Fig. 6 Decay times of neutron pulses.

Figure 6 shows the decay times of the exponential functions fitted to the decay as a function of wavelength.

Discussion and Conclusions

We have found that composite moderators of liquid Hydrogen offer good prospects for providing high intensities and short pulses in high power pulsed spallation neutron sources. These are the first results on composite cold moderators. The measurements reported here probably do not represent a system that would actually be employed in practice, however, optimization within the framework of this idea will certainly lead to systems with better performance. Further work along this line is certainly justified. Meanwhile, the present results are a valuable benchmark against which to test calculations and to compare with other alternative moderator systems.

Intensities and to a lesser extent the spectra and pulse shapes of moderator systems depend sensitively on the relative arrangement of moderators, primary source(s) and reflectors. As systems become more complex, the problem of providing consistent normalization for different systems in different geometries becomes more difficult. We cannot resolve the question of how to present data so that results are precisely transferable from one geometry to another, but believe that the present results are reasonably representative of the performance of composite moderators in other source arrangements.

References

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