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Composite Hydrogen-Solid Methane Moderators

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ABSTRACT

This paper describes the results of Monte-Carlo calculations for a coupled moderator on a low-power pulsed neutron spallation source and is part of the design study for a second target station for the ISIS spallation source. Various options were compared including hydrogen, solid methane, grooving the solid methane and compound moderators made of hydrogen in front of solid methane. To maximise the neutron current at low energies two strategies appear to emerge from the calculations. For instruments that view a large area of moderator surface a layer of hydrogen in front of a thin solid-methane moderator is optimum, giving a gain of about a factor 10 relative to the current liquid hydrogen moderator on the existing ISIS tantalum target. For instruments that only view a restricted area higher flux, corresponding to a gain of 13.5, can be achieved with the use of a single groove or re-entrant hole in the moderator.

1. INTRODUCTION

This study was carried out in connection with the proposed second target station on the ISIS pulsed neutron source at the Rutherford Appleton Laboratory, in the UK.

The existing target station utilizes a 200μA, 800MeV proton beam operating at a repetition frequency of 50Hz. The planned extension to the source will involve increasing of the proton current to 300μA and constructing a second target station, optimized for the production of cold neutrons. One pulse in five will be diverted to the second target station, which will operate at a proton current of 60μA and a repetition frequency of 10Hz.

The relatively low power (48kW) of the second target station can be put to advantage. In particular, a solid methane moderator, similar to those used on the Intense Pulse Neutron Source at Argonne in the US, would be a practicable alternative to liquid para-hydrogen moderators. However, the problems with the storage of radiation damage in solid-methane and the potential for the explosive release of this energy means that great care has to be taken with the design and operation of solid-methane moderators. With appropriate pre-moderation the heating rate could be reduced sufficiently to enable the moderator to be annealed only once per day.

As currently envisaged, the Second Target Station will include two moderators: a coupled moderator for instruments that are relatively insensitive to pulse widths, and a decoupled moderator for instruments that require short pulse. This paper focuses on design and materials options for the coupled moderator, for which the main optimization criterion is the maximization of neutron intensities, particularly in the low-energy range (0 to 5meV). Pulse widths and time distributions are a secondary consideration, but should be taken into account because they may be important for some instruments. Two moderator materials are considered: liquid para-hydrogen and solid methane, either singly or in combination.

This paper explores two basic options with the aim of significantly improving upon the performance of a simple flat-faced solid methane or liquid hydrogen moderator: The first option involves the use of grooves in solid methane, permitting the instrument to view the cold, intense flux in the moderators' interior. The second option involves the use of a compound moderator in which solid methane is put behind the hydrogen. The hydrogen compensates for the hardening of the leakage spectrum from the surface of the methane, and the methane compensates for the gap in the hydrogen scattering law at low energies. Above 15meV the moderator performance and time distributions of the compound moderator are similar to hydrogen. At lower energies the results depend on the hydrogen thickness, a reasonable choice for which is about 5cm which enhances the neutron intensity by 50% relative to solid methane while producing similar pulse widths.

In Section 2, we discuss the properties of these materials. Solid methane is a typical moderator material, whose total cross-section rises with decreasing energy. However, liquid para-hydrogen is unusual; its neutron cross section is relatively high at energies above the para-ortho transition energy (14.7meV), but it is comparatively transparent below this energy. This changes the way these materials perform and the way they can be used as moderators.

In section 3, we describe the basic parameters of the Monte-Carlo calculations, including details of the simplified target/moderator/reflector geometry. In sections 4, 5 and 6 we present the results of calculations for flat-faced moderators, multiply grooved moderators and singly grooved moderators respectively. Finally, in section 7 we summarise the overall conclusions of the study.

2. THE PROPERTIES OF SOLID METHANE VERSUS HYDROGEN

The two moderator materials under consideration are liquid hydrogen at approximately 20K and solid methane, which would in practice be run at about 26-27K in order to ensure safe operating conditions (the higher the temperature the fewer the annealing cycles). The hydrogen density in the two materials is quite different: approximately 0.076 atoms per barn cm for solid methane but only 0.042 for liquid hydrogen! At neutron energies above about 100meV this is the most important difference, but atomic binding effects become significant at lower energies. Figure 1 shows the total cross sections for liquid ortho and para-hydrogen and solid methane. The most striking feature of these curves is the unusual behaviour of the para-hydrogen cross section, which almost vanishes below the ortho-

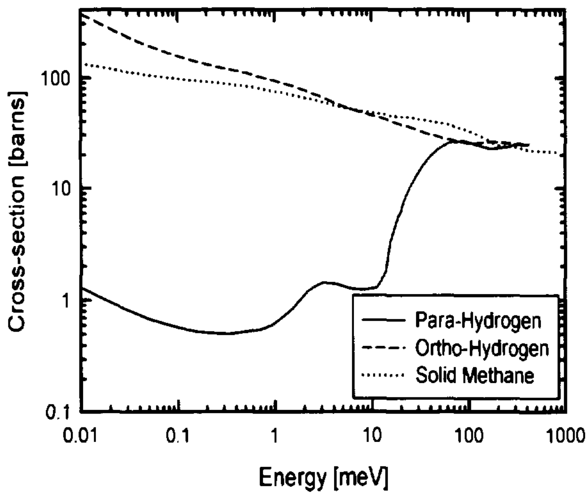

Figure 1. The cross-sections of ortho and para-hydrogen (dashed and solid line resp.) and solid methane (dotted line).

para transition energy. Liquid hydrogen moderators on spallation sources are believed to exist in an almost pure para-hydrogen state, due the action of powdered magnetic catalysts in the cooling circuit, and are therefore relatively transparent to neutrons below 15meV (although it should be noted that a small proportion of ortho-hydrogen would significantly increase the neutron cross section in this energy range). The low-energy properties of liquid para-hydrogen are in fact highly advantageous. The rapid release of sub-15meV neutrons from the moderator produces relatively sharp time distributions and high levels of neutron leakage in this energy range, although the leakage drops away at lower energies.

Both moderators have advantages and disadvantages: solid methane is an excellent moderator, but the neutron's mean free path is very short at low energies so the optimum width is narrow (2 to 3cm). Making the moderator thicker means that you get larger flux at its centre, but does not increase the surface leakage.

3. THE DETAILS OF THE CALCUALATIONS

All the calculations were carried out using the Monte-Carlo code MCNPX [1], which is a new version of MCNP[2] which incorporates the capabilities of LAHET[3] into the MCNP code, thus permitting the calculation to include high-energy charged particles (e.g. protons, pions) as well as neutrons, photons and electrons.

The geometry used is illustrated in Figure 2. This is a highly simplified version of the currently envisaged geometry for the proposed ISIS Second Target Station. The reflector is an 80cm cube of heavy-water cooled beryllium, represented by a homogenized mixture (20% D_2O, 80% Be by volume). The target is a tungsten cylinder of diameter 6cm, surrounded by a 10cm square cell containing light water (with the dual function of cooling the target and pre-moderating the neutron spectrum to reduce the heat load on the solid methane moderators to acceptable levels). The source was a proton beam of energy 800MeV and current 60μA. The beam diameter was 3cm, and the radial intensity profile was a Gaussian with σ = 0.5cm and a 3σ cut-off. Only one moderator was included in the configuration. This was placed above the target in 'wing' geometry, with beam ports on both sides. The thickness and composition of the moderator

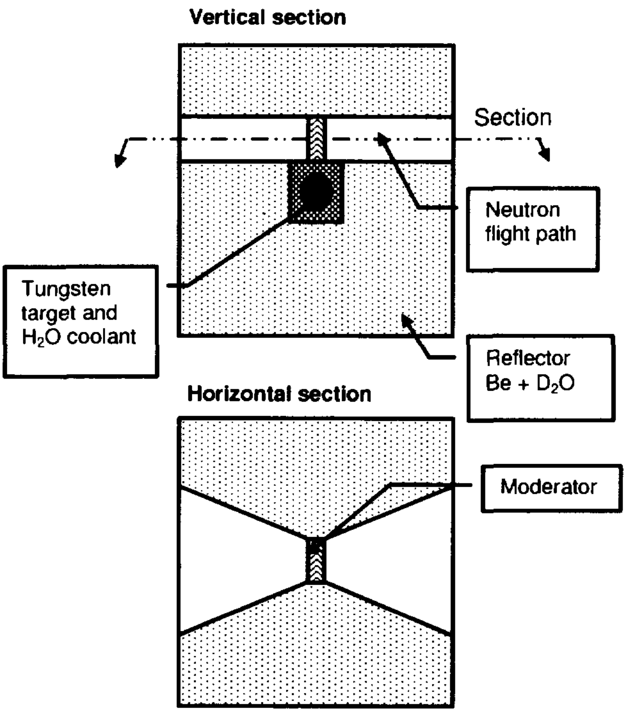

Figure 2. The simplified geometries used for the calculations: a) A vertical section showing the cylindrical target and coolant/pre-moderator cell, the slab moderator and its associated neutron beam ports, and the reflector. And b) a horizontal section showing the slab moderator, its beam ports and the reflector.

varied between calculations but the transverse dimensions were always 12 × 12 cm^2.

The thermal neutron cross-section kernels [4] used in this study were those made available to participants in the Cold Moderator study. Documentation on the methods used to describe the kernels is given in Ref. [5].

Two methods were used for the tallying of the neutron current at the moderator surface. The time-integrated current was obtained, for a direction orthogonal to the moderator surface, using a distant point detector. The time distribution of the current was obtained using a surface-crossing tally, but the time-dependent leakage was normalized to produce the time-integrated values determined by the point detector.

4. THE OPTIMISATION OF THE FLAT FACE MODERATORS

Three series of calculations were carried out:

(i) Solid methane moderators of variable thickness (in 1cm steps up to 6cm), tallies scored for both faces.

(ii) Liquid hydrogen moderators of variable thickness (in 1.6cm steps up to 9.6cm), tallies scored for both faces.

(iii) Composite moderators consisting of a hydrogen moderator in front of a methane moderator. Tallies were scored for the hydrogen face only.

Table 7. The time-integrated intensities in the 0-5meV energy range. The results are given relative to a 4.8cm thick liquid hydrogen moderator. The lateral dimensions of the moderator are 12 x 12 cm, and the temperatures are 26K and 20K for the solid methane and hydrogen respectively. The error in the last significant figure is shown in brackets. The time integration is from 0 to 30,000μs.

Methane thickness(cm)	*Hydrogen thickness (cm)* 0.0	1.6	3.2	4.8	6.4	8.0	9.6
0	0.0	0.464(2)	0.768(3)	1.000(4)	1.197(5)	1.357(6)	1.491(6)
1	1.140(5)	1.618(6)	1.84(1)	1.98(1)	2.08(1)	2.15(1)	
2	1.351(6)	1.74(1)	1.92(1)	2.06(1)	2.14(1)		
3	1.326(6)	1.693(9)	1.90(1)	2.03(1)	2.12(1)		
4	1.285(6)	1.661(9)	1.84(1)	1.98(1)			
5	1.241(6)	1.610(9)	1.82(1)				
6	1.214(6)						

The results for time-integrated leakage are shown in Table 7. All values are given relative to a reference case: 4.8 cm hydrogen. It is interesting to compare the non-composite hydrogen and methane moderators. The methane moderator has an optimum thickness of between 2 cm and 3 cm, whereas the hydrogen moderator continues to give a better performance as its thickness is increased. However, it is clear that the composite moderator show a significant performance enhancement relative to non-composite moderators at energies below 15 meV.

Figure 3 shows the calculated FWHM pulse widths for non-composite moderators and for composite moderators with 2 cm methane. The pulse widths from methane are largely independent of thickness, whereas, at low energies, the hydrogen pulse widths rise. However, the hydrogen moderator still gives sharper pulses than the methane for thicknesses of less than 10cm, albeit with a flux penalty.

The results for the composite moderator are more complex; with a switch between different behaviours above and below 15meV. At low energies the pulse width get narrower when a thin layer of hydrogen is added, but the addition of further hydrogen significantly increases the pulse widths as well as increasing the neutron leakage. At higher energies, the moderator starts looking like a methane moderator but becomes more hydrogen like as the hydrogen thickness increases. A good compromise would be about 2 cm methane with 5 cm hydrogen, giving a pulse width about the same as the pure methane moderator but with a performance gain of over 2 relative to 4.8 cm hydrogen and 50% better than 2 cm methane alone.

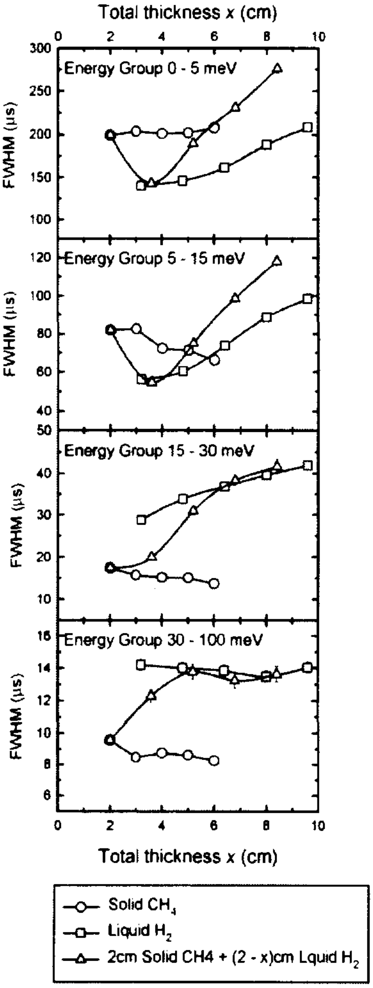

Figure 3. Plots of the FWHM pulse widths versus the moderator thickness, for both the flat face moderators, in all four of the calculated energy ranges.

Figure 4 shows the time distributions from 4.8 cm and 6.4 cm hydrogen moderators, and for some of the cases with 2cm solid methane. It is clear that increases in the hydrogen thickness also increase the pulse width, but the bulk of the increase lies within the pulse; there is no large increase in the tail. The composite moderator shows that increasing the amount of hydrogen increases both pulse width and height with no detrimental effect on the tail.

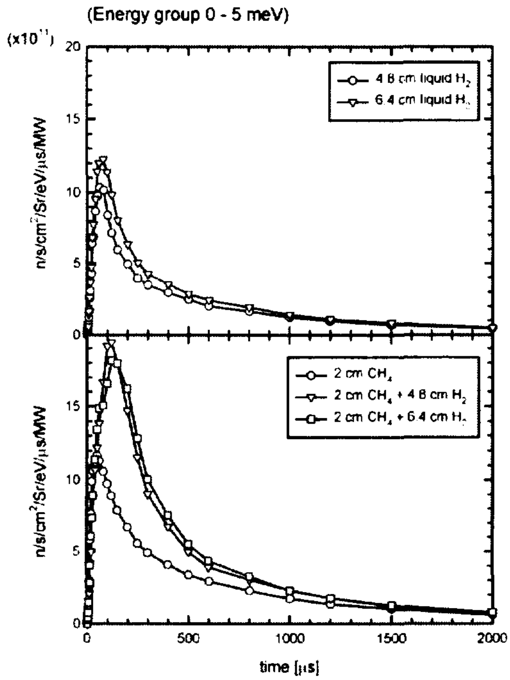

Figure 4. The pulse shapes for the 0 to 5meV energy group for some of the simple and composite moderators flat face moderators.

5. THE OPTIMISATION OF MULTIPLY GROOVED MODERATORS

One of the common ways to improve the flux from solid methane is to add a series of narrow grooves into the moderator. This presents us with a huge parameter space to study, so we have selected a few key points in this space to give us an idea about the possible performance. For the purposes of this study, we have added four horizontal grooves (Figure 5). In the initial configuration, the grooves extended half way into the moderator; the groove height was specified as 1.5 cm, with 1.5 cm of solid methane between the grooves. However, the enlargement of the grooves was found to be desirable, and later calculations used 2cm grooves separated by 1cm of moderator material. Similarly, the extension of the grooves further into the moderator proved to be advantageous, and later calculations all used grooves whose depth was 2/3 of the moderator thickness.

Two options for the use of para-hydrogen in conjunction with a grooved methane moderator were explored: adding hydrogen inside the grooves, and/or an additional flat-faced hydrogen moderator in front of the grooves.

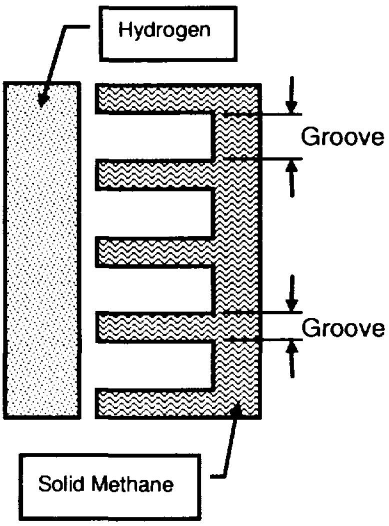

Figure 5. The geometry of the multiply grooved moderators (vertical section). The moderator has lateral dimensions of 12 x 12 cm with four horizontal grooves. Various geometries have been run with different groove widths, separations and depths. Compound moderators incorporating a slab of hydrogen moderator in front of the grooves were also investigated.

Results for the time-integrated current are shown in Table 8 for selected cases. The best results appear to be from the case with hydrogen in front of empty grooves. However, this is only the case if the time integration is taken over the whole of the long tail. A more careful look at the peak shapes (figure 7) shows that for empty grooves the shape is double peaked and that adding hydrogen to the grooves adds height to the peak and produces a clean single shape with 10% extra flux in the 0 to 2000 μs region. Controlling the long time tail is obviously something that needs to be addressed, perhaps by adding a poisoning layer deep in the reflector.

In Figure 6 the FWHM of these selected cases is shown as a function of thickness. One of the remarkable results is how the addition of hydrogen into the grooves sharpens up the pulse. From Figure 7 we can see that above 200μs the peak shapes for hydrogen in front of empty grooves and grooves filled with hydrogen are roughly the same (although the empty grooves are very slightly higher), but below 200μs there is a higher, sharper peak.

Table 8: A summary of calculated neutron intensities from multiply grooved moderators. The results are given relative to a 4.8cm flat-faced liquid hydrogen moderator. The lateral dimensions are 12 x 12 cm and the temperatures are 26K and 20K for the solid methane and hydrogen respectively. The time integration is from 0 to 30,000μs.

Methane thickness	*Hydrogen thickness*	*Groove geometry* *Height*	*Separation*	*Fractional depth*	*Groove material*	*Relative intensity*
	(cm)					
8.0	0.0	1.5	1.5	0.5	void	1.621(9)
8.0	0.0	2.0	1.0	0.5	void	1.705(9)
8.0	0.0	1.5	1.5	0.67	void	1.692(9)
8.0	0.0	2.0	1.0	0.67	void	1.81(1)
2.0	0.0	2.0	1.0	0.5	void	1.307(8)
4.0	0.0	2.0	1.0	0.5	void	1.588(9)
6.0	0.0	2.0	1.0	0.5	void	1.644(9)
8.0	0.0	2.0	1.0	0.5	void	1.71(1)
10.0	0.0	2.0	1.0	0.5	void	1.72(1)
2.0	0.0	2.0	1.0	0.67	void	1.164(6)
4.0	0.0	2.0	1.0	0.67	void	1.623(9)
6.0	0.0	2.0	1.0	0.67	void	1.76(1)
8.0	0.0	2.0	1.0	0.67	void	1.81(1)
10.0	0.0	2.0	1.0	0.67	void	1.83(1)
2.0	0.0	2.0	1.0	0.67	H_2	1.410(8)
4.0	0.0	2.0	1.0	0.67	H_2	1.762(9)
6.0	0.0	2.0	1.0	0.67	H_2	1.82(1)
8.0	0.0	2.0	1.0	0.67	H_2	1.81(1)
10.0	0.0	2.0	1.0	0.67	H_2	1.79(1)
6.0	1.6	2.0	1.0	0.67	void	2.01(1)
6.0	3.2	2.0	1.0	0.67	void	2.14(1)
6.0	4.8	2.0	1.0	0.67	void	2.21(1)
6.0	1.6	2.0	1.0	0.67	H_2	1.98(1)
6.0	3.2	2.0	1.0	0.67	H_2	2.05(1)
6.0	4.8	2.0	1.0	0.67	H_2	2.12(1)

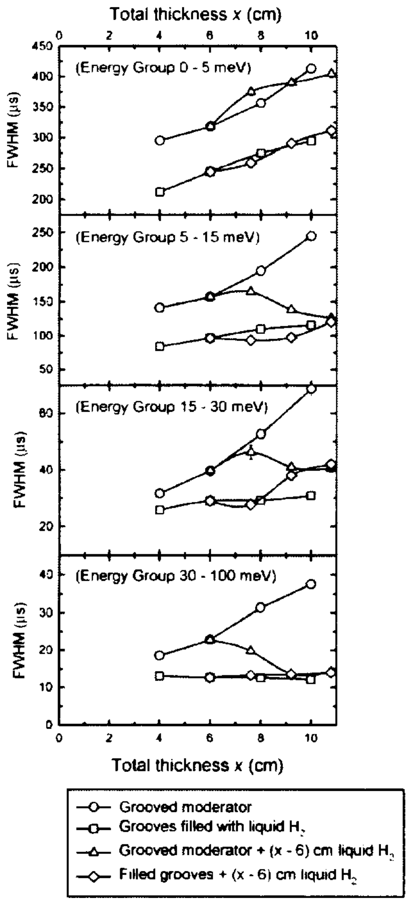

Figure 6. Plots of the FWHM pulse widths versus the moderator thickness for the multiply grooved moderators. There are four grooves, each 2cm high and separated by 1cm of solid methane. The methane moderator is 6cm thick with grooves 4cm deep and has lateral dimensions 12x12 cm.

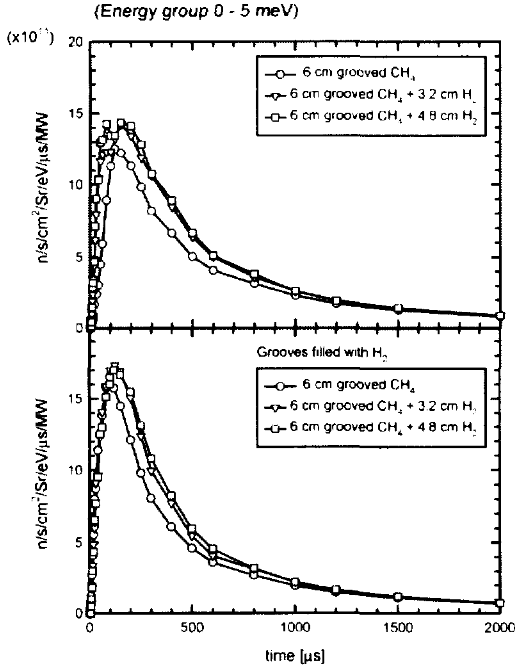

Figure 7. A comparison of time distributions for some of the multiply grooved moderators in the energy group 0 to 5meV.

6. SINGLE GROOVE CALCULATIONS

Some neutron instruments, such as small angle or reflection machines only view a restricted area of the moderator face. This makes it possible to either use a large single groove or re-entrant hole in the moderator, which offers the prospect of larger gains than either the flat face or multiply grooved moderators

Figure 8 shows the geometry of the single-groove moderator, the design of which had been optimized in previous studies. A tapered slot of height 4cm, depth 6.5 cm and width 8.4 cm (at rear) is cut into a moderator of dimensions 10 cm (height) . 16 cm(width) . 11 cm (depth).

The results for the time-integrated neutron current (again normalized to a simple 4.8cm hydrogen moderator) and pulse widths can be seen in table 3. The 0 to 5meV energy group shows a gain of approximately 2.7 relative to a simple 4.8cm hydrogen slab, and represents a significant improvement on the compound and multiply-grooved cases. However, it should be noted that the performance of the single-groove moderator is inferior to the flat-faced and multiply grooved moderators at energies above 5meV.

Figure 9 shows the calculated time distributions in the 0 to 5meV energy group, compared with the optimal compound-moderator case (4.8cm liquid hydrogen in front of 2cm solid methane). The grooved moderator produces a somewhat longer pulse width than the compound flat-faced moderator. However, it is evident that the gain from the grooved versus

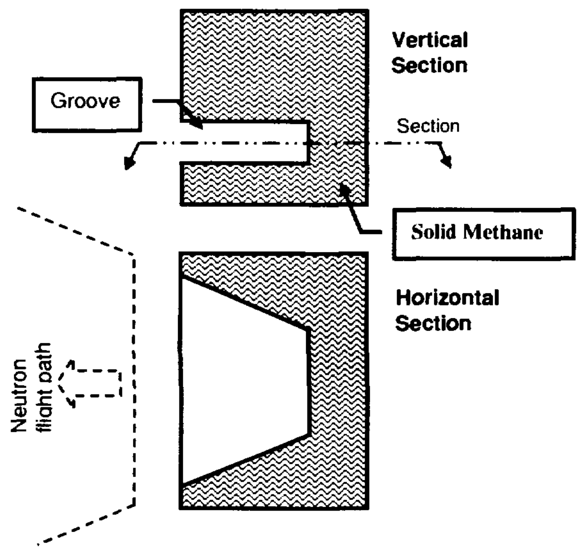

Figure 8. The geometry of the single groove (vertical section and horizontal sections). The groove is positioned at the base of the moderator at the peak of the internal flux. The groove is in fact a tapered recursive hole. It is 4cm high and the taper is keyed into the shape of the neutron flight path. The moderator is 10 cm high, 16 cm wide and 11cm deep.

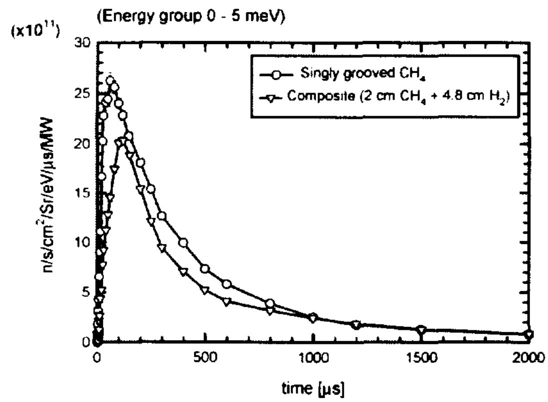

Figure 9. The pulse shape from the singly groove moderator both compared to the optimum flat-face composite moderator (2cm CH_4 + 4.8 cm H_2).

the flat-faced moderator is 'real' in the sense that the bulk of the increased neutron flux occurs near the peak and not in the long-time tail.

Table 2. Calculated neutron intensities and pulse widths for the single-groove moderator.

Group (meV)	*Relative Intensity*	*FWHM of pulse (μs)*
0 - 5	2.70(1)	251(8)
5 - 15	1.281(9)	70(4)
15 - 30	0.618(6)	11.5(8)
30 - 100	1.02(1)	7.6(5)

7. DISCUSSION

Except where otherwise stated, the following discussion will focus on the moderator performance in the 0 to 5meV energy group relative to a simple 12 x 12 x 4.8cm^3 liquid hydrogen moderator.

In the case of the flat-faced compound moderators; good results were obtained using a liquid hydrogen moderator in front of a 2cm thick solid methane moderator. The performance increased with the thickness of the hydrogen moderator, at the expense of increasing pulse widths. Our design brief for the coupled moderator was for the highest possible intensity at low energies. The moderator which best fits this brief is the combination of 2cm of solid methane and 4.8 cm of hydrogen; giving a performance gain of 2.06 relative to a simple 4.8 cm hydrogen slab.

The concept of a multiply grooved methane moderator also proved highly successful. From the point of view of maximizing neutron intensity, the best arrangement proved to be a

4.8 cm hydrogen slab in front of a 6cm grooved methane moderator (groove depth 4 cm, height 2 cm, separation between grooves 1cm, no material in the grooves). Flowing hydrogen into the grooves produced significantly 'cleaner' and sharper pulses. The best compromise is a 3.2 cm hydrogen moderator in front of a 6cm solid methane moderator (grooves as described above but containing hydrogen), which would give a gain of 2.05 relative to a simple slab. Given that this gain is very similar to that for the simpler flat face design it is unlikely that this will be chosen in reality.

The results for a single-groove moderator were good, the optimum result being a gain of 2.70 relative to a simple 4.8 cm hydrogen slab. This would provide a substantial performance enhancement for cold-neutron (< 5 meV) for instruments which view a limited section (~4cm) of the moderator height.

It is interesting to compare the results with the liquid hydrogen moderator on the existing ISIS target (on the basis of flux per MW of beam power.) In the case of the flat-faced moderator (4.8cm hydrogen, 2cm solid methane), the calculated gain relative to the liquid hydrogen moderator on ISIS is 20.7 and the gain for the single groove moderator is 27.0. These gains sound very large but two points should be remembered:

a) The hydrogen moderator on the existing target is decoupled.
b) These gains are exaggerated by the omission of significant system components (moderator cans, target pressure vessel and cooling manifold) from the simplified model used in this paper. Later calculations in a more realistic geometry have shown that the simplified geometry provides accurate estimates of relative moderator performance, but over-predicts the absolute fluxes by about a factor of two; thus the true gains for the flat-faced and singly grooved moderators are 10 and 13.5 respectively.

7.1 The optimum moderator for instruments view the whole moderator

The optimum cases for the flat-faced and multiply grooved compound moderators give almost identical performance for the time-integrated neutron current. However, the flat-faced configuration is considered superior to the multiply grooved case for three reasons:

a) The flat-faced moderator gives a sharper pulse (FWHM 231μs) compared with the grooved case (FWHM 291μs).
b) Given that the flux gains are similar, the extra engineering complexity of the multiply grooved moderator does not appear to be warranted.

Group (meV)	***Relative Intensity***	***FWHM of pulse (μs)***
5 + 2 + 5cm 'sandwich' ($H_2 + CH_4 + H_2$)		
0 to 5	1.944(8)	234(3)
5 to 15	1.458(6)	92(1)
15 to 30	0.884(4)	39.8(8)
30 to 100	0.840(8)	13.4(3)
4.8 + 2cm compound moderator ($H_2 + CH_4$) (viewing the hydrogen face only)		
0 to 5	2.06(1)	231(4)
5 to 15	1.538(8)	98(2)
15 to 30	0.941(5)	38(1)
30 to 100	0.921(9)	13.6(5)

Table 4. A summary of neutron intensities and pulse widths from the 'sandwich' moderator.

Table 4 summarizes the pulse widths and moderator performance for a 5 + 2 + 5 cm sandwich as described above, compared with the 4.8 + 2cm flat-faced compound moderator. The sandwich produces almost identical time distributions to the two-part compound moderator, but there is a small penalty of about 7% in the time-integrated intensity.

7.2 The optimum moderator for instrument that view a restricted area of the moderator

Energy Range (meV)	*Relative Intensity*	*FWHM of pulse(μs)*
	Grooved face	
0 - 5	2.65(2)	241(7)
5 - 15	1.264(8)	64(3)
15 - 30	0.623(6)	11.4(8)
30 - 100	1.04(1)	7.7(5)
	Compound flat face	
0 - 5	1.95(1)	209(3)
5 - 15	1.345(7)	83(2)
15 - 30	0.754(5)	35(1)
30 - 100	0.780(8)	13.2(4)

Table 5: Summary of the neutron intensities and pulse widths from a single groove moderator with a compound flat face.

Table 5 summarizes the pulse widths and moderator performance for a modified version of the single-groove moderator, in which 2cm of solid methane is removed from the flat face and replaced with 4cm of liquid hydrogen. This design successfully combines the performance of both the single-groove and flat-faced moderators. This will allow those instruments that can to take advantage of the high flux from the groove, but also have a high flux face available for those instruments that require a view of the full moderator height. This design has been adopted as the basis for future studies.

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