Pulsed Neutron Source Cold Moderators
Concepts, Design and Engineering

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ABSTRACT

Moderator design for pulsed neutron sources is becoming more and more an interface area between source designers and instrument designers. Although there exists a high degree of flexibility, there are also physical and technical limitations. This paper aims at pointing out these limitations and examining ways to extend the current state of moderator technology in order to make the next generation neutron sources even more versatile and flexible tools for science in accordance with the users' requirements.

1. INTRODUCTION

Neutrons are usually freed from nuclei in a process called nuclear evaporation and are released with kinetic energies corresponding to a Maxwell distribution with a characteristic temperature of some 2×10^{10} K, i.e., a few MeV. In order to shift their energies to where they are useful for condensed matter research, a process called "moderation" is applied.

While neutron moderation in nuclear fission reactors is a necessity in order to maintain the chain reaction and therefore very limited choices exist in designing and engineering the moderator, this is not true for accelerator driven neutron sources. This opened up a whole new field of research aimed at trying to understand in detail the moderation process with respect to both its temporal and spatial characteristics. The goal is to make moderator design especially for pulsed sources an integral part of the design of neutron scattering instruments and allow the users to specify their needs up front. Instrument designers have, in fact become increasingly aware of this opportunity and are beginning to give more and more expert input to source designers in terms of their needs. In this context a language has developed in which moderators are characterized by their temperature, indicating the characteristic energy of the spectral distribution they generate, and by "high intensity" or "high resolution", depending on whether the emphasis is on the highest possible integral intensity per pulse or on the shortest possible pulse duration. Clearly, this is a rather crude simplification with realities being much more sophisticated. Nevertheless it provides a useful means to give an overview of the trends. Table 1 lists the results of two evaluations made for the European Spallation Source (ESS, 5 MW([1]) or the US National Spallation Neutron Source (NSNS; 1-4 MW) [2].

While both projects envisage of the order of 40 instruments, none of them opts for high intensity ambient temperature moderators (which would be the easiest ones to provide) and only a few instruments request high resolution ambient temperature moderators. Clearly the emphasis on both sources is on moderators below room temperature and, taking intermediate temperature and cryogenic moderators together, the consensus is strikingly good. A possible explanation why intermediate temperature moderators are not on the ESS-list (in fact, one such request was forwarded but didn't show up in the final list) might be that intermediate temperature moderators are traditionally associated with liquid methane, which is known to cause serious problems at high radiation levels. In any case, the requirements listed in Table 1 are reason enough to revisit the opportunities and limitations in cold moderator engineering and to search for novel solutions to old problems.

Table 1. Instrument requests to moderators for ESS[1] and NSNS[2] Numbers before and behind the / refer to the high repetition and low repetition rate target stations respectively.

		ESS: 50 Hz / 10 Hz	
	Ambient temperature		Cryogenic (25 K)
High intensity	0 / 0		0 / 8
High resolution	9 / 0		15 / 12
		NSNS: 60 Hz / ≤20 Hz	
	Ambient temperature	intermediate temp. (100 K)	Cryogenic (25 K)
High intensity	0 / 0	0 / 0	0 / 9
High resolution	4 / 0	11 / 6	1 / 6

Slowing down of neutrons in a medium occurs by elastic collisions (elastic in a nuclear physics sense, i.e., without conversion of energy into mass; energy is, of course, exchanged between the neutrons and the atoms they collide with). Therefore, it is intuitively clear that the following conditions must be fulfilled for a substance to make a good moderator for cold neutrons:

1. the energy transfer per collision must be high;
2. the cross section must be high to have a high collision probability;
3. the macroscopic density of moderator atoms per unit volume must be high to have a short time between collisions;
4. there must be suitable modes at low energies for the neutrons to couple to;
5. neutron absorption, which increases inversely with neutron velocity, should not be too serious;
6. it must be possible to remove the heat from the moderator efficiently to maintain low temperature conditions throughout; and
7. the moderator material should have sufficient chemical stability under irradiation.

2. TIME STRUCTURE OF SPALLATION NEUTRON SOURCES

Being driven by an accelerated proton beam, spallation neutron sources can be designed with almost any desired time structure on the fast neutron flux emerging from the target, provided there is not too much moderation in the target itself. Liquid metal targets, as they are now widely considered in the new medium to high power spallation sources fulfill this condition in an almost ideal way. Depending on the type of accelerator driving the source three classes can be distinguished:

2.1 Continuous Spallation Sources

For spallation neutron sources fed from an accelerator with no macro-time structure such as a cyclotron or a cw-linac the optimization goal naturally is a high time average flux. Use of low absorption materials in and around the target is crucial in order to ensure a long lifetime of moderated neutrons. The moderator should be large to achieve complete thermalization and minimum leakage losses. Typically, a 2 m diameter D_2O tank will surround the target, acting as moderator and reflector at the same time. Cold moderators in such sources will be of the re-thermalizing type, i.e. they will be embedded in the surrounding D_2O and will shift the ambient temperature equilibrium spectrum to cryogenic temperatures. This is the same situation as it exists in modern research reactors. The only existing source of this type is SINQ [3,4] at the Paul Scherrer Institut in Switzerland. An account on the first operating experience with the SINQ cold moderator is given in [5].

2.2 Short Pulse Neutron Sources

While continuous neutron sources favor those neutron applications that require a high time average flux, there is a whole class of neutron scattering techniques that work in a pulsed mode (time of flight experiments) and hence can be served best by a high peak flux at appropriate repetition rates. It is with those instruments in mind that short pulse spallation neutron sources were developed. Their characteristic feature is that they generate fast neutron pulses which are short enough so they don't contribute noticeably to the width of the moderated neutron pulse in the whole energy interval of interest. This requires single turn extraction of a stored beam from a ring that can either be a rapid cycling synchrotron, as in IPNS, KENS, and ISIS or an accumulator/storage ring fed by a full energy linac as in LANSCE. Both types are also being proposed in new projects. The optimization criterion in this type of source is a short pulse duration without disproportionate losses in peak flux. Rapid slowing down and a short neutron life time in the moderator after thermal equilibrium has been reached are at a premium. In order to accomplish the latter, moderators are often surrounded by neutron absorbing material (decouplers) to prevent slow neutrons from being returned from the reflector. Absorbing material is even placed at a certain depth inside the moderator to limit the time slow neutrons spend diffusing out of the moderator (Figure 1a). This kind of arrangement has been designated "high resolution" in Table 1. Obviously, this is on the expense of intensity. Up to a factor of 10 or more can be gained by doing away with the decoupler and poison and using "coupled" moderators where the extra pulse width can be tolerated from a resolution point of view. This type of moderator is referred to as "high intensity" in Table 1. For short proton pulses neutron pulses from both moderators have a steep rising edge and comparable (within a factor of 2) peak flux. The extra intensity for coupled moderators is primarily in the longer trailing edge.

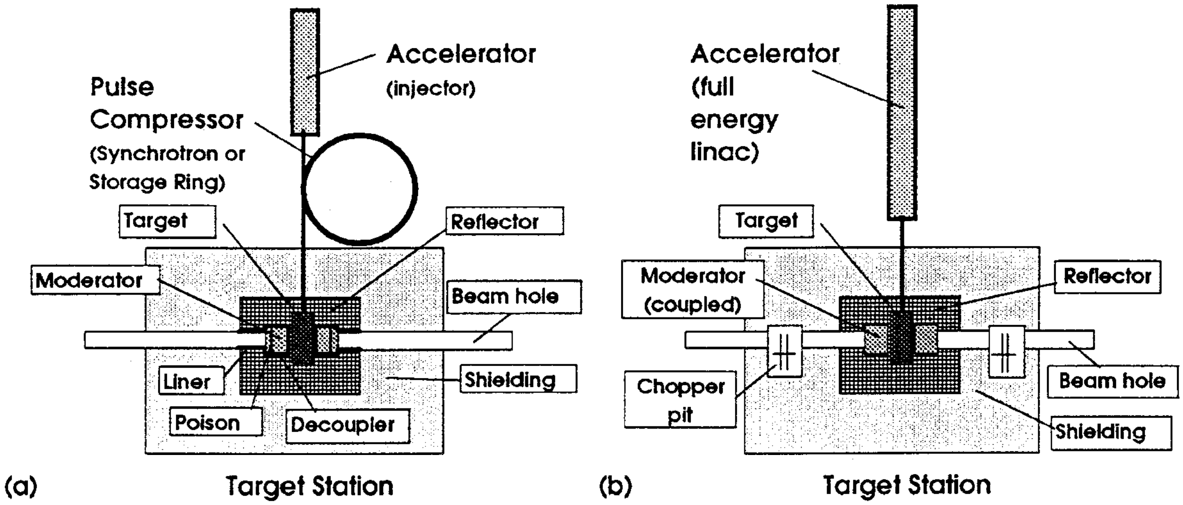

Figure 1. Schematic representation of pulsed neutron source arrangements. (a) short pulse source with decoupled and poisoned moderator (b) long pulse source with coupled moderator.

2.3 Long Pulse Spallation Neutron Sources

A spallation source fed directly from a pulsed linac is usually referred to as a long pulse source, meaning that the pulse length of the proton beam is significantly longer than the time it takes the neutrons to decay in the moderator. The pulse width of these sources is thus essentially determined by the duration of the proton pulse and will be of the order of 1 ms. Again, high time average flux is an important optimization criterion, but also the pulse shape becomes important, especially if chopper instruments for time of flight spectroscopy are to be used. This becomes immediately obvious from the pinhole effect of a chopper with short opening time near the moderator, as it is needed for a broad band of transmitted neutrons. As can be seen from Figure 2, the neutron intensity at the sample position varies as a function of time not only due to the spectral modulation (see below) but also reflects the shape of the moderator pulse. Precise knowledge of the intensity as a function of wavelength is crucial in inverted time of flight experiments because data taken at different wavelengths must be compared to each other intensity wise. It is, therefore, important in this type of source to find the right compromise between high peak flux and neutron life time in the moderator [6]. [For this reason the moderators, even cryogenic ones, will still be of the slowing-down type but no measures will be taken to artificially reduce the neutron life time, i.e. the moderators will be tightly coupled (Figure 1b). A moderator design as used in continuous sources would distort the pulse shape too strongly due to the very long life time of the neutrons. To a good approximation the pulse shape in a long pulse source can be represented for all neutron energies by the relations

$$\Phi(t) = \Phi_{as}(1-\exp(-t/\tau)) \qquad \text{for } t \le t_p$$

$$\Phi(t) = \Phi_{as}(1-\exp(-t_p/\tau))\exp(-(t-t_p)/\tau) \qquad \text{for } t \ge t_p \qquad (1)$$

with t_p being the duration of the proton pulse and τ the life time of the neutrons which depends on the neutron energy (see below). Φ_{as} is the flux that would be reached if the beam was on at full intensity all the time. The average flux is $\Phi_{av} = \Phi_{as}\, t_p \nu$ with ν being the pulse repetition rate. Frequently more than one τ will be required to characterize a moderator in its environment.

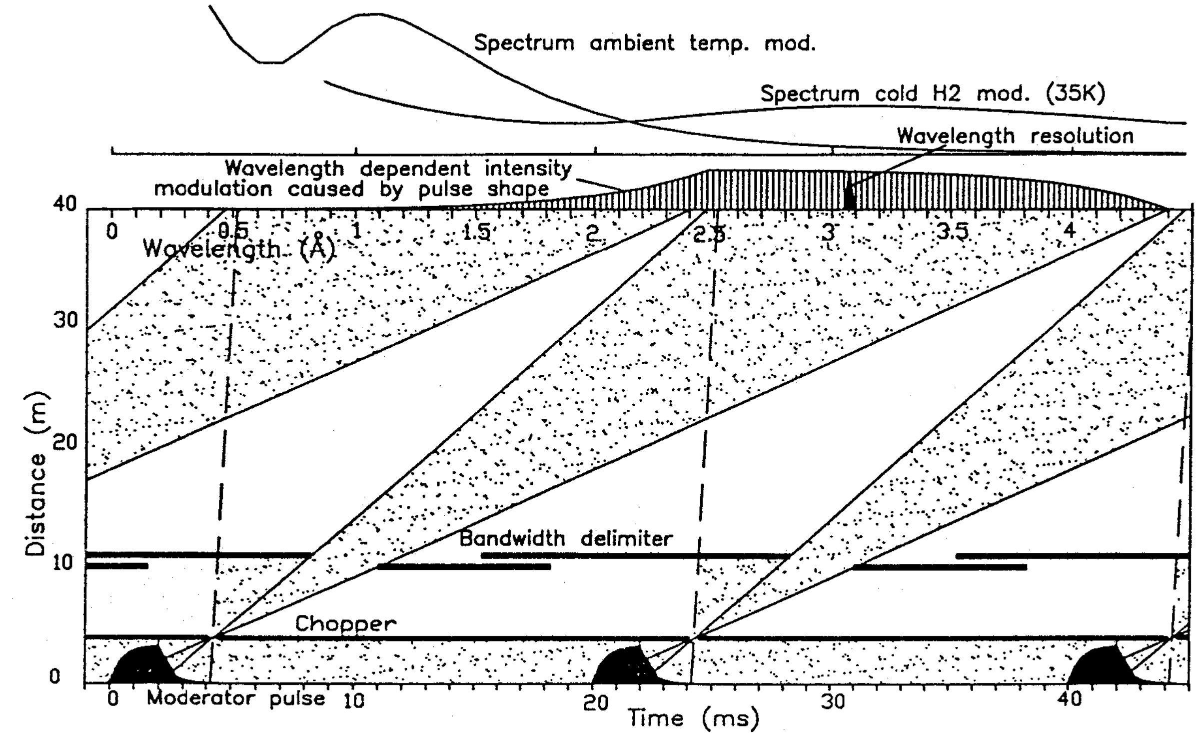

Figure 2. The pinhole effect from a chopper close to the moderator imposing an additional modulation on the neutron spectrum at the sample, on top of the natural spectral distribution.

Although the first long pulse neutron source, SNQ [7] was proposed almost two decades ago, no such facility has so far been realized. However, recently interest in long pulse sources is growing again, with particular emphasis on the use of slow neutrons (cold moderators) [8], requiring a longer pulse duration than planned for SNQ.

3. THEORETICAL BACKUP: SLOWING DOWN OF FAST NEUTRONS IN MODERATORS

Although Monte Carlo methods are now widely used to study complex target-moderator-reflector assemblies as a unit, we briefly review, in a cursory way, the mechanisms and processed involved in neutron moderation in order to illustrate the above conditions (1) through (4) and to give a clue on the directions to head for.

In each collision during the slowing down process the neutron transfers, on average, a certain fraction of its kinetic energy to the atoms of the moderator, i.e. E_1/E_2 is a constant. The logarithm of this constant, $\xi = \langle \ln(E_1/E_2) \rangle$ is called the logarithmic energy decrement and is given by

$$\xi = \ln(E_1/E_2) = 1\text{-}(\alpha_0\varepsilon/(1\text{-}\alpha_0\varepsilon)) = 1 \text{ for } A\text{=}1 \text{ and} \approx 2/(A\text{+}2/3) \text{ for } A>1 \qquad (2)$$

with $\varepsilon = \ln(1/\alpha_0)$ and $\alpha_0 = (A\text{-}1)^2/(A\text{+}1)^2$

A is the atomic number of the moderator atom. This immediately shows that hydrogen is the most efficient element in terms of neutron moderation. The number x of collisions required to slow down a neutron from its original energy E_0 to an energy E_f is then given by

$$x = (1/\xi)\ln(E_0/E_f) \qquad (3)$$

Values of ξ and x (for E_0 = 2 MeV and E_f = 1 eV) are listed in Table 2, together with other quantities relevant for neutron slowing-down for selected elements and compounds.

The value of the free atom scattering cross section σ_{fr} given in Table 2 is more or less constant over a large energy range for most elements, as long as the neutron energy is higher than or of the order of the binding energy of the atoms in molecules or condensed matter. This is the case down to around 1 eV. As an example the scattering cross section of hydrogen is given in Figure 3.

The probability of a collision to happen depends on the macroscopic cross section

$$\Sigma = N\sigma = 1/\Lambda \qquad (4)$$

with σ being the microscopic cross section and N the number of atoms per unit volume. Λ is the mean free path between collisions. Λ being constant, the time t_i between collisions and therefore the number of neutrons present in a certain velocity interval is inversely proportional to the neutron velocity, v_i.

$$t_i = \Lambda/v_i = 1/(\Sigma_{fr} v_i) \quad \text{or} \quad v_i t_i = \Lambda \approx \text{const.} \qquad (5)$$

Because the neutron flux can be interpreted as the product of the number of neutrons per unit volume in a certain velocity interval times their velocity, the neutron flux in the slowing-down regime depends on the neutron velocity as v^{-2} or as E^{-1}. In small moderators this law will be modified. A convenient representation of the neutron current from the moderator surface reads [9]

$$<I(E)_{sd}> = I(E_0)(E_0/E)^{1-\alpha} = [EI(E)]\big|_{E0}\,(1/E)(E/E_0)^{\alpha} \qquad (6)$$

with E_0 being a reference energy (usually 1eV) and α depending on absorption in and leakage from the moderator during the slowing-down process. For non-reflected moderators α is of the order of 0.2, but can be significantly reduced, depending on the moderator environment. $[EI(E)]\big|_{E0}$ is frequently referred to as “epithermal beam current” I_{epi}.

Table 2. Quantities relevant for neutron slowing down for a few elements and hydrogen rich compounds

	Element									
Parameter	H	D	Be	C	N	O	Fe	Ni	Hg	Pb
A	1	2	9.01	12.01	14	16	55.85	58.71	200.6	207.19
σ_{fr} (10^{-24} cm^2)	20.51	3.40	6.18	4.73	10.03	3.75	11.21	17.89	26.53	11.01
σ_b (10^{-24} cm^2)	82.02	7.64	7.63	5.551	11.51	4.232	11.62	18.5	26.8	11.118
σ_{inc}	80.27	2.05	0.0018	0.001	0.5	0.008	0.4	5.2	6.6	0.003
σ_a (10^{-24} cm^2)(*)	0.3326	0.0005	0.0076	0.0035	1.9	0.0002	2.56	4.49	372.3	0.171
ρ (g/cm^3)(*) (+)	0.07	0.163	1.85	2.3	0.804	1.13	7.9	8.9	13.55	11.3
N ($10^{24}/cm^3$)	0.042	0.049	0.124	0.115	0.035	0.043	0.085	0.091	0.041	0.033
$\Sigma_c = N\sigma_c$ (cm^{-1})	0.86	0.17	0.76	0.55	0.35	0.16	0.96	1.63	1.08	0.36
ξ	1.000	0.725	0.206	0.158	0.136	0.120	0.035	0.034	0.010	0.010
γ	1.000	0.584	0.143	0.108	0.093	0.082	0.024	0.023	0.007	0.006
x (2MeV→1eV)	14.5	20.0	70.3	92.0	106.5	121.0	410.0	430.8	1460.1	1507.9
vt_s (cm)	3.47	21.37	13.58	24.51	44.28	108.83	59.86	36.77	187.06	576.49
$v\Delta t_s$ (cm)	1.74	10.69	6.79	12.25	22.14	54.42	29.93	18.39	93.53	288.24
$v\Delta t_{1/2}$	3.47	24.81	19.01	34.88	63.47	156.85	88.73	54.54	279.66	861.96

(*) σ_a is for thermal neutrons (2200 m/s); cross section data are from [10].
(+)for H, D, N, O,CH_4 and NH_3 ρ is given for the liquid at the respective boiling points at 1 atm.

Table 2 Cont'd.

	Compound †					
Parameter	H_2O	CH_4	$(CH_2)_n$	NH_3	TiH_2	D_20
A	18	16	14	17	49.92	20
σ_{fr} (10^{-24} cm^2)	44.8	86.8	45.7	71.5	42.43	10.5
σ_b (10^{-24} cm^2)	168.3	333.6	169.6	257.6	163.3	19.5
σ_{inc}	160.5	321.1	160.5	241.3	165.5	4.1
σ_a (10^{-24} cm^2)(*)	0.665	1.334	0.669	2.898	6.755	0.001
ρ (g/cm^3)(*) (+)	1	0.453	0.94	0.682	3.978	1.1
N ($10^{24}/cm^3$)	0.033	0.017	0.040	0.024	0.048	0.033
$\Sigma_c = N\sigma_c$ (cm^{-1})	1.50	1.48	1.85	1.73	2.04	0.35
ξ	0.926	0.954	0.913	0.879	0.968	0.510
γ	0.923	0.951	0.908	0.873	0.967	0.405
x (2MeV→1eV)	15.7	15.2	15.9	16.5	15.0	28.4
vt_s (cm)	2.11	2.09	1.72	1.89	1.505	13.51
$v\Delta t_s$ (cm)	1.054	1.046	0.861	0.945	0.753	6.754
$v\Delta t_{1/2}$	2.16	2.13	1.78	1.97	1.52	16.85

† For the compounds cross sections and N are given per molecule.

The result $v\Delta t_S = const.$ *is very important for pulsed neutron sources for two reasons:*

(a) It shows that, if neutrons from the slowing-down regime are used, the resolution of a time of flight instrument, which is inversely proportional to the neutron velocity, remains constant because the pulse width decreases as the velocity increases.

(b) While the time-average flux of slowing-down neutrons on a pulsed source is not higher than on a continuous source, rather lower because of the faster moderators used, the instantaneous flux during the pulse is enhanced by a factor v on top of the compression into short pulses.

This is why there is a strong incentive to use cryogenic moderators to shift the Maxwellian distribution (see below) to lower velocities and thus extend the slowing-down regime.

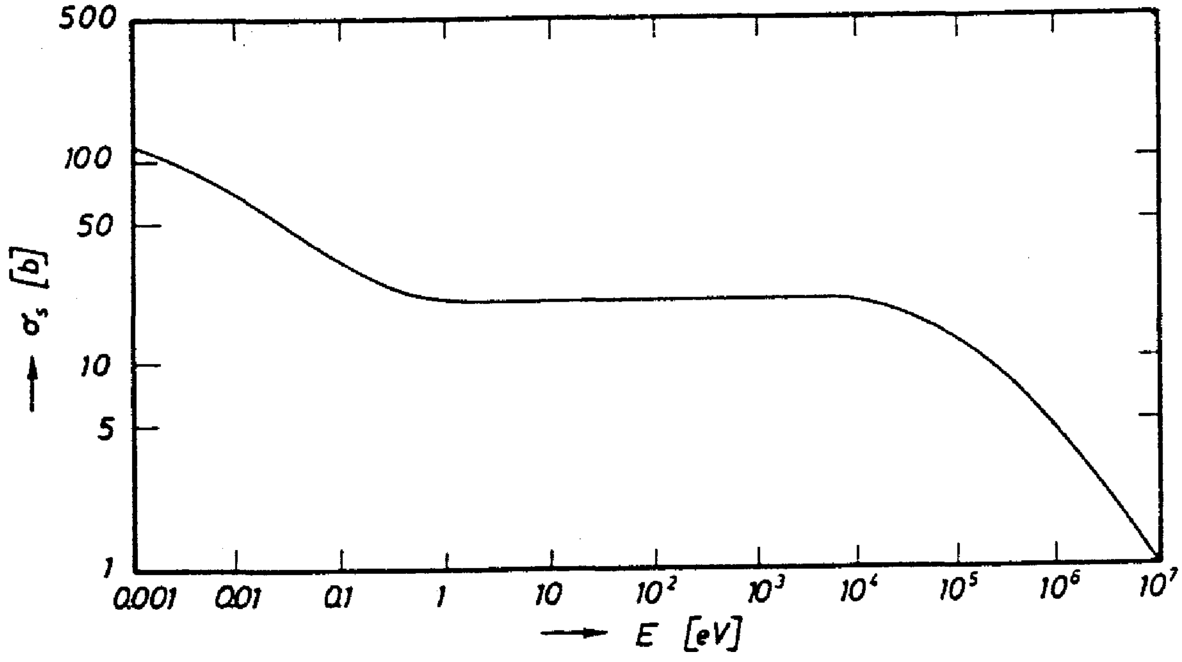

Figure 3. The scattering cross section of hydrogen as a function of the energy of the incident neutron.

Equation 5 also suggests that the total time it takes to slow a neutron down to a final velocity v is inversely proportional to v. A more rigorous theory [9] yields for the time dependence of the slowing down flux following a short source pulse a function of the product vt only:

$$\Phi(v,t) \sim (\xi\Sigma_{fr} vt/\gamma)^{2/\gamma} \exp(-\xi\Sigma_{fr} vt/\gamma) \qquad (7)$$

$$\text{with } \gamma = 1 \text{ for } A=1 \text{ and } \approx 4/(3A) \text{ for } A>1. \qquad (8)$$

The average slowing-down time t_S, its standard deviation Δt_s and the FWHM $\Delta t_{1/2}$ are then found as:

$$vt_s = (1+2/\gamma)\gamma/(\xi\Sigma_{fr}) \qquad (9)$$

$$v\Delta t_s = (1+2/\gamma)^{1/2}\gamma/(\xi\Sigma_{fr}) \qquad (10)$$

$$v\Delta t_{1/2} = 3/(\xi\Sigma_{fr}) \qquad (11)$$

t_s gives the delay between when the peak of the velocity distribution reaches v in the moderator (t_0 for the time of flight measurement) relative to when the proton pulse was injected into the target, whereas Δt_s and $\Delta t_{1/2}$ represent the pulse width. Numerical values for these quantities are included in Table 2 for some elements and for compounds with high hydrogen content. In cases where the substances are gaseous at room temperature, values corresponding to the liquid at its boiling temperature are given. The quantities ξ and γ were computed using the formulae given in [9] rather than the approximations of equations 2 and 8, but differences are minor even for the light elements. Averaging for compounds was done by weighting by the free atom cross sections σ_{fr}.

Clearly materials to be used in an optimized moderator-reflector system should have a large value of Σ_{fr}. In the case of the moderator also a large value of ξ is desirable, whereas for a good reflector ξ should be small in order to return as many neutrons as possible to the moderator without significant energy loss for a short pulse length. It is obvious from the data in Table 2 that, as an element, hydrogen is by far the best moderator. In order to have a high enough density, it must be either in the liquid (or supercritical) state, or a compound with high hydrogen content must be used. If any of the hydrogen-rich compounds can be used, for which $v\Delta t_s \approx 1$cm, this makes a better moderator than liquid hydrogen with $v\Delta t_s = 1.74$ cm. We will come back to this question in the chapters below.

Although the approximation of a constant scattering cross section is valid only above about 1 eV, the slowing-down regime extends to an energy corresponding to roughly five times the effective moderator temperature, T_{eff}, because moderation continues until equilibrium with the thermal motion of the moderator atoms is reached. Below this limit an energy distribution of the neutrons is observed, which represents more or less thermal equilibrium between the moderator atoms and the neutrons. It is described by the Maxwellian distribution:

$$\Phi_M(E) = \Phi_{th}\, E/(k_B T_{eff})^2 \exp(-E/(k_B T_{eff})) \tag{12}$$

where Φ_{th} is the thermal neutron flux and $k_B = 0.086165$ meV/K is Boltzman's constant. The transition between the slowing-down regime and the thermal equilibrium spectrum which at about $5kT_{eff}$ is usually taken into account by a switch function $\Delta_1(E)$, by which the slowing down spectrum is multiplied. A frequently used formula is

$$\Delta_1 = 1/(1+5\, k_B T_{eff})^5 \tag{13}$$

The full representation of the spectrum, including equation 6 then reads:

$$\Phi(E) = \Phi_{epi}(1/E)(E/E_0)^{\alpha}\, \Delta_1(E) + \Phi_{th}\, E/(k_B T_{eff})^2 \exp(-E/k_B T_{eff}) \tag{14}$$

Note that, while Φ_{th} is an energy integral, Φ_{epi} is not, although they have the same physical dimension. Φ_{epi} corresponds to what is called flux per unit lethargy $\Phi(u)$ in reactor terms, with $u = \ln(E_0/E)$.

Figure 4 shows how the peak of the neutron distribution shifts to lower energies as the moderator temperature is reduced. The E^{1} dependence of the spectrum is clearly seen in the measured data above 1 eV. Below 1 eV the effect of a cadmium sheet inserted in the moderator to reduce the life time and hence pulse width causes some flux depression, but the extension of the slowing-down regime to lower energies is obvious. Although there is no true thermal equilibrium in this kind of a moderator, a Maxwellian distribution is still a reasonably good description for the low energy part.

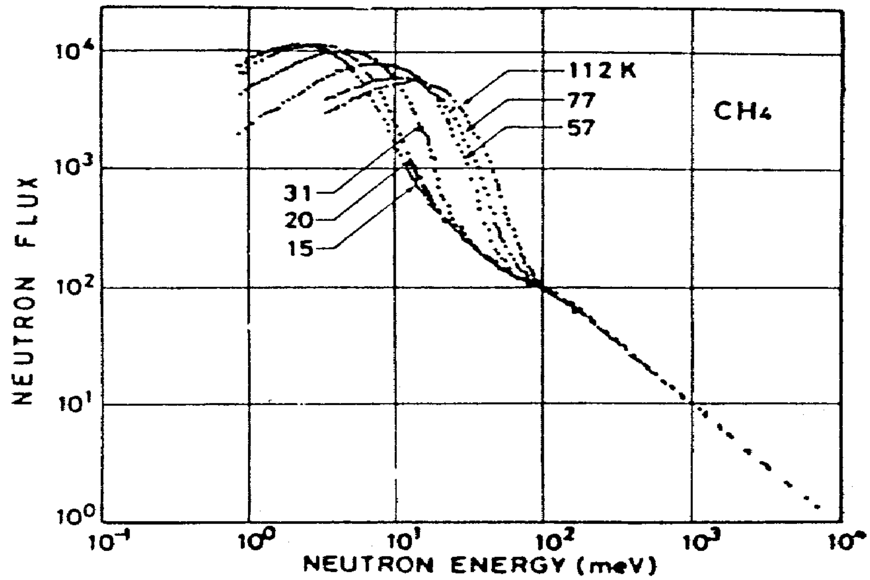

Figure 4. Energy spectra from a methane moderator at various temperatures [11].

In the thermal equilibrium regime the number density of moderator atoms is not as important as it is in the slowing down regime, but attention must be paid to the scattering cross section as well as to neutron absorption. As the neutron energy drops below the binding energy in condensed matter, atoms can no more be knocked out of their positions, but energy must be transferred to elementary excitations for further slowing-down. This is where the properties of condensed matter become important. At the same time the scattering cross section changes gradually from its value for the free atom, σ_{fr}, to that for the rigidly bound state, σ_b. The two are related to one another by

$$\sigma_b = \sigma_{fr}\ ((A+1)/A)^2 = 4\ \pi\ b^2 \qquad (15)$$

Values for σ_b are included in Table 2. b is called the bound atom scattering amplitude or scattering length and represents the phase shift the neutron wave experiences in the scattering process. Since the interaction is not with individual nuclei anymore but with the whole assembly of nuclei, the distinction between coherent and incoherent scattering becomes important. Because coherent interaction means that the phases of all scattered neutrons are the same, it is the average of their scattering amplitudes that enters into Equation 15 for coherent scattering. Equation 15 then reads:

$$\sigma_{inc} = 4\ \pi\ \langle b\rangle^2 \qquad (15a)$$

On the other hand, the mean total scattering cross section of an assembly of nuclei is the average of their cross sections, $4\ \pi\ \langle b^2\rangle$. Therefore, the incoherent scattering is given by:

$$\sigma_{coh} = 4\ \pi\ (\langle b^2\rangle - \langle b\rangle^2) \qquad (15b)$$

The distinction between coherent and incoherent scattering is important in the context of neutron moderation, because in crystalline solids the coherent scattering cross section vanishes for neutron momenta smaller than $4\pi/d_{max}$ with d_{max} being the largest lattice spacing in the crystal. In an almost purely coherent scatterer (such as, for example D_2O) neutrons with lower energies than this "Bragg cutoff" pass through the material practically unscattered, whereas above the Bragg cutoff intense elastic scattering would prevent neutrons from penetrating the moderator. (D_2O-ice would, therefore, not make a good cryogenic moderator, even if the heat could be removed. For liquid D_2O the cross section does not go all the way to the incoherent value for small energies, as shown in Figure 5, because there is no long range correlation of the atomic positions.) In a predominantly incoherent scatterer, such as H_2O, correlation effects are almost not visible in the total scattering cross section (Figure 5).

There can be various reasons for the occurrence of incoherent scattering, such as a mixture of isotopes with different scattering amplitudes or nuclear spin. The latter is particularly important for hydrogen: Interaction of a neutron with spin 1/2 and a nucleus with spin J can happen via compound nuclei with spin (J+1/2) or (J-1/2). The corresponding scattering amplitudes are generally different and are designated as $b+$ and $b-$, respectively. Hydrogen, being a molecule of two atoms with spin 1/2, can have both spins parallel ($J = 1$, ortho hydrogen,) or antiparallel ($J = 0$, para hydrogen). Values for $b+$ and $b-$ are 10.817 and -47.420 fm respectively, which enter in different combinations to give for the cross section *per atom*:

$$\sigma^{H}_{par} = 3.13\times10^{-24}\ \mathrm{cm}^2 \quad \text{and} \quad \sigma^{H}_{ort} = 98.2\times10^{-24}\ \mathrm{cm}^2.$$

While at room temperature about 75% of the hydrogen gas is in the ortho state, nearly all molecules convert to the para state at low temperatures in an unperturbed system. This leads to an energy dependent cross section of hydrogen at low temperatures as shown in Figure 6.

For deuterium (D_2) at low temperatures the ortho state dominates. Its cross section is

$$\sigma^{D}_{ort} = 18.6\times10^{-24}\ \mathrm{cm}^2.$$

3. PRACTICAL ISSUES

The high cross section and large energy transfer per collision clearly make hydrogen the most attractive atomic species for pulsed source (cold) moderators. As a consequence of the peculiarities of the cross section in the bound state and the discrete energy levels in condensed matter and molecules, Equations 13 and 14 do not describe the spectra obtained, e.g., from liquid hydrogen or methane correctly. However, for the present purpose we will not reiterate on these details any further.

While liquid hydrogen does not suffer from radiation damage, has sufficient low lying energy modes, and is liquid down to 14 K, allowing easy heat removal, it is of moderate number density only and hence yields relatively wide pulses (Table 2). Nevertheless, practically all cryogenic moderators on medium or high power neutron sources use liquid or supercritical hydrogen (or liquid deuterium).

Methane has a rich spectrum of low energy modes (Figure 7a) due to nearly free rotational transitions of the tetrahedral molecule, the lowest one lying at 1 meV. Relative to liquid hydrogen solid methane at 20 K yields in excess of 3 times more cold neutrons for wavelengths of 4 Å and longer (Figure 8).

Polyethylene (Figure 7b) and TiH_2, which look even better than methane in terms of pulse width, have no low lying modes and hence, like ice, might be good down to a few meV (Figure 9).

A common problem to all hydrogen rich compounds is the fact that they are solid at the temperature of liquid hydrogen. This has two important consequences:

1. heat removal must be through conduction rather than convection / evaporation as in the case of a liquid, which is difficult due to the low thermal conductivity at cryogenic temperatures and
2. structural radiation damage as well as the products of radiolysis and other radiation effects are stored in the volume and can react exothermally when the system warms up slightly, sometimes in a catastrophic way. Details of this process are given in [13].

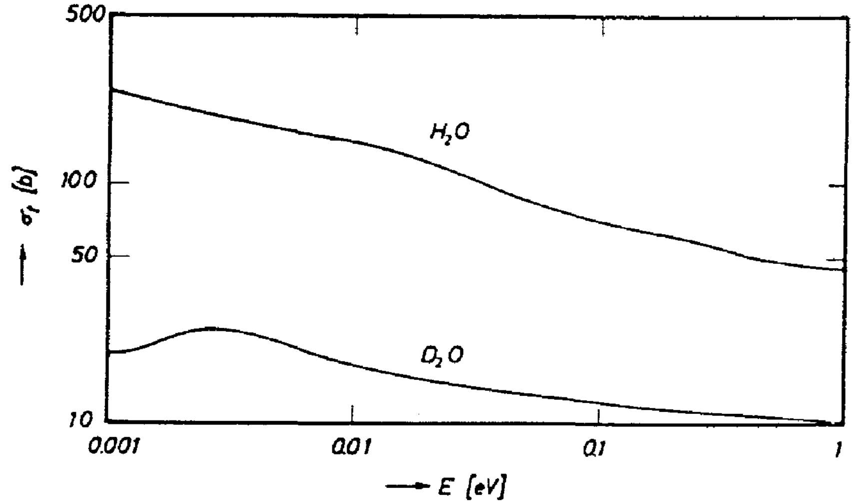

Figure 5. Cross section of light and heavy water as a function of neutron energy below 1 eV.

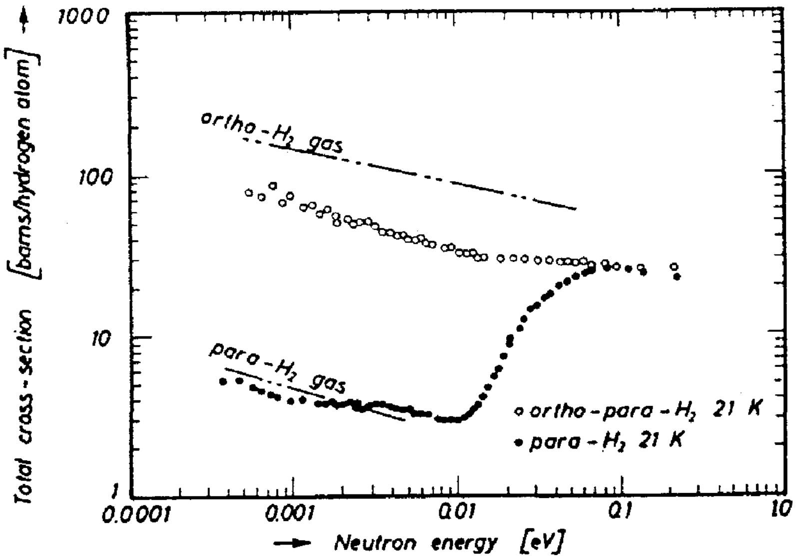

Figure 6. Scattering cross section of hydrogen at low temperature as a function of neutron energy.

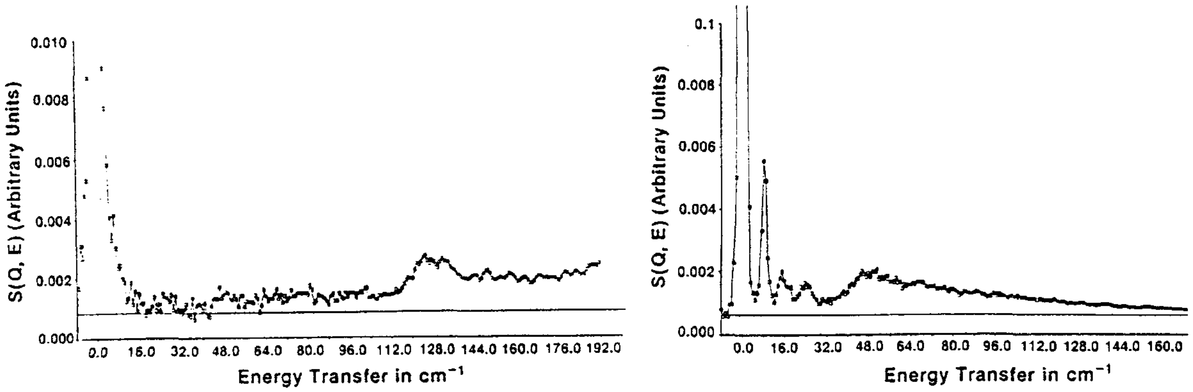

Figure 7. Cross section as a function of neutron energy for polyethylene at 12 K (left) and methane at 9 K (right). The scales are the same in both panels.

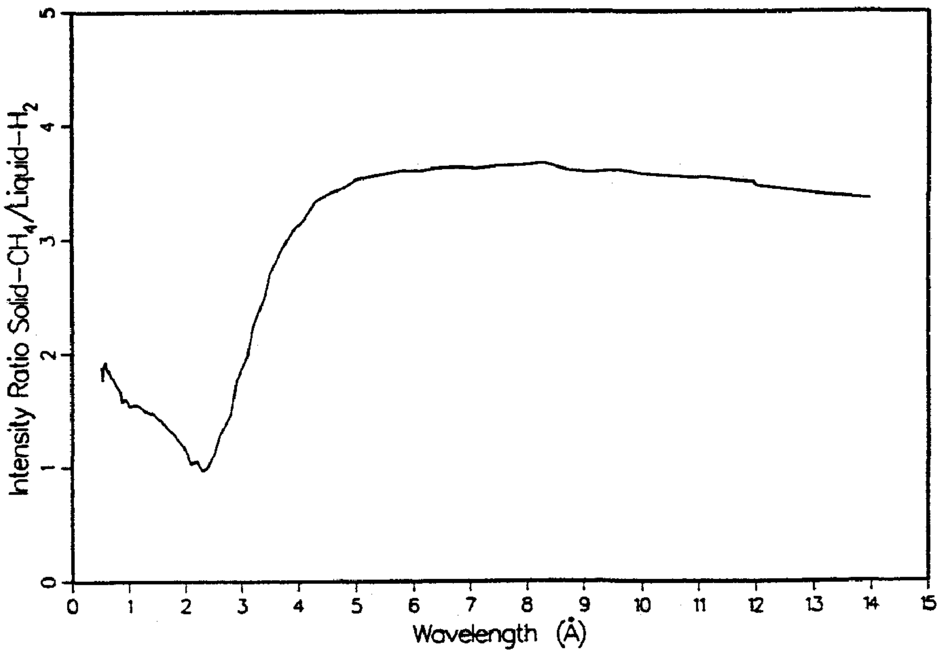

Figure 8. Ratio of the spectral intensities of solid CH_4 and liquid H_2 moderators. The same moderator was used in both cases, and all geometry was the same [12].

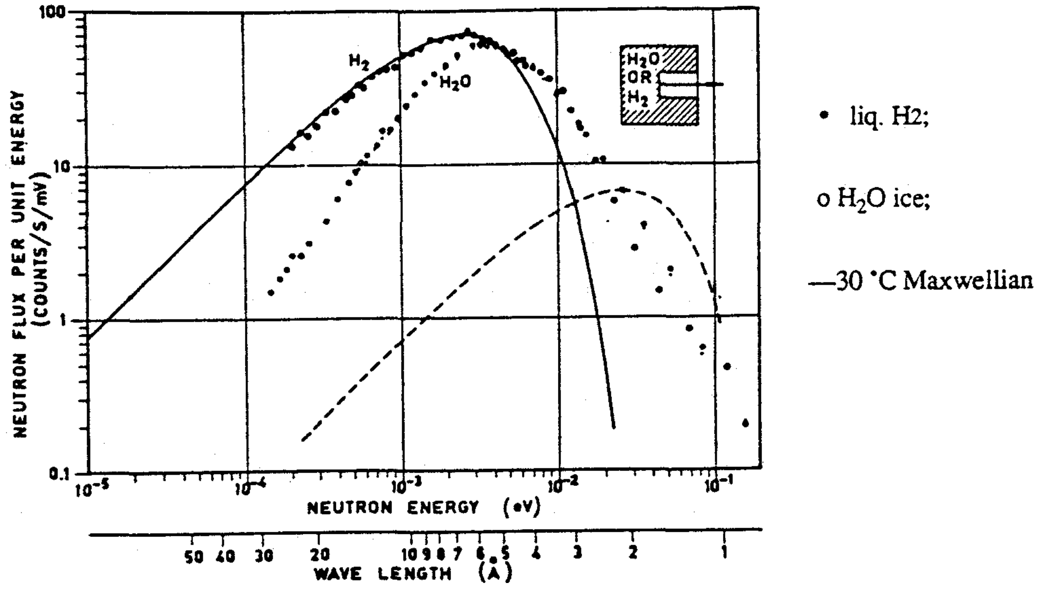

Figure 9. Neutron spectra obtained from H_2 and ice at 20 K.

For solid methane this effect that has led to the destruction of the moderator container in the past. In order to avoid this problem, solid methane moderators must be warmed up periodically [13,14]. In any case, due to problems of heat removal, massive solid methane moderators can only be used on low power spallation sources.

Methane that solidifies at 90.7 and boils at 109.2 K (at 1 atm) would make an almost ideal intermediate temperature moderator in liquid form. But it was found that radiolysis is a serious problem [15] because polymerization products generated tend to cling to the walls of the moderator container and to plug the system sooner or later.

4. NEW ROUTES

It is clear from the foregoing discussion, that solid moderators offer a variety of opportunities both at very low temperatures and in the intermediate temperature range around 100 K, if the problems of heat removal and stored energy release can be solved. One way to accomplish this was discussed in [16] with special emphasis on the use of methane. In order to

- achieve high hydrogen content in the volume at very low temperature (performance aspect)
- allow safe and continuous removal of polymerization products even at high radiation levels (operational aspect), and to
- devise a system where the incidental spontaneous release of stored energy would not affect the whole volume (safety aspect),

a concept was considered in which pelletized moderator material (methane) is closely packed in the moderation volume and is continuously moved through the system at a rate that avoids excessive damage while irradiated. Heterogeneous cooling would be provided by a fluid flowing between the pellets. A conceptual sketch of the system is shown in Figure 10.

Removing the pellets from the moderation volume by mechanical means independent of the feeding mechanism allows to control the pellet transport rate and the coolant flow separately and thus optimize the residence time of the pellets in the moderator vessel. As the pellets cross the moderator vessel, heat removal from their volume is by conduction only and their size will have to be chosen such that the equilibrium temperature gradient from the center to the surface remains within tolerable limits. The problem here is the low thermal conductivity of most materials at low temperatures. If this condition cannot be fulfilled, the residence time in the moderator volume will have to be rather short. However, this would have to be feared only under fairly extreme conditions. Estimates given in [16] show that, for spherical methane pellets of 4-7 mm diameter and a volumetric heating rate of 1 W/cm^3, equilibrium temperatures in the center would be 10-30 K above the surface temperature and would be reached after 1-2 min. it is, therefore, likely that proper cooling of the pellets can be achieved and that the residence time will be dominated by radiation damage. It should be noted that spontaneous release of stored energy in individual pellets need not be a problem, as long as there is no avalanche effect across the whole volume. Even then, consequences would be much less serious than in a massive solid due to the porosity of the pack.

While the system outlined in Figure 10 is designed for re-circulation of the moderator material as well as of the coolant, this is, of course, not a necessity. Depending on what material is used for moderator and how long it can reside in the moderator vessel, a once through system may be more efficient and economic.

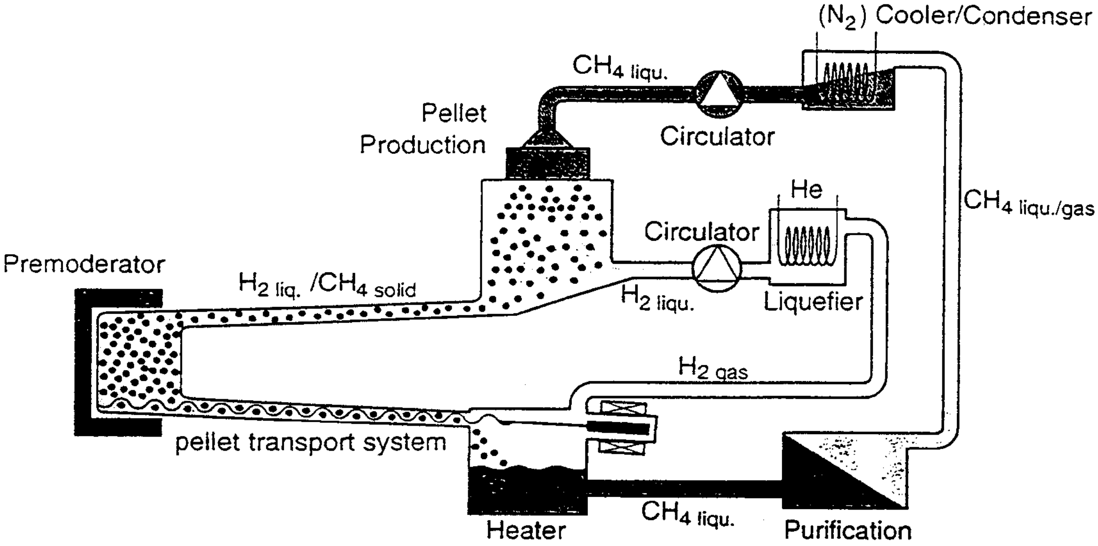

Figure 10. Pelletized methane moderator system with mechanical control of the pellet removal rate from the moderation volume [16].

Also, while methane seems to be the preferred material for very low temperatures according to what was said above, there is no conclusive evidence that it is the only choice. For example, silane, SiH_4 (T_m = 88.2 K) or stannic hydride, SnH_4 (T_m = 124 K) might have similar low lying energy modes and so might other polyatomic substances. A systematic study is required, including not only the energy spectrum but also other relevant properties to assess the existing options.

In the intermediate temperature range, where very low-lying modes are less important, there exist a variety of substances, including ice, which might be good candidates for moderator materials. Cooling may either be by a liquid such as nitrogen or, possibly, cold hydrogen or helium gas. In the case of nitrogen formation of dangerous nitrogen-oxygen mixtures under irradiation has been reported, but it should be possible to eliminate this risk.

Although most of this is speculative at the present stage, it might be worth noting, that one could also conceive using porous materials, such as zeolites, microporous or mesoporous titania etc, loaded with the moderator material to act as a carrier and to improve heat removal. Some of these materials are known to be able to store hydrogen at densities up to several times that of its liquid state. Another interesting observation in this context is that of very low energy modes in hydrogen loaded nanocrystalline metals which are ascribed to hydrogen in the grain boundaries.

In view of the need to improve the performance and design flexibility of cold moderators, especially for pulsed neutron sources, the ESS-team decided to set up a task group that will explore the opportunities outlined above with the goal to devise a workable system by the time the final design of ESS will be decided.

The following is a list of topics to be dealt with and a rough working plan:

- Analyze candidate materials for pellets
  - CH_4 and homologues, other organic compounds, H_2O, NH_3, metal hydrides...(?)...
  - Hydrogen density
  - absorption
  - radiochemistry
  - thermal properties
  - mechanical properties
  - etc.
- Analyze density of states at low energies
  - literature
  - neutron scattering
  - other
- Select cryogenic fluids
  - temperature range
  - behavior under irradiation
  - compatibility with materials
- Investigate methods for “pellet” fabrication
  - direct freezing
  - use of porous materials
  - others (?)
- Develop method for mass transport
  - pellets, using “easy” varieties (PE, ice...)
  - fluid (pumping system, pressure levels ..)
- Determine optimum “pellet” size
  - coolability
  - fabrication
  - transport
  - coolant pressure drop

- Build test rig (verify calculations and concepts)
- Carry out neutronics experiments (AGS, COSY) to verify calculations
- Test in existing neutron source (SINQ, Risø (?))

If successful, the outcome of this study has potentially significant impact on the final design of the ESS target station, depending on whether the pellet transport must be horizontal as shown in Figure 10, or whether a vertical removal of the pellets from the moderator vessel can be accomplished.

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