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The ACoM Collaboration An International Effort to Develop Advanced Cold Moderators

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Abstract

Several laboratories world wide have decided to join forces in developing a cryogenic moderator system that would come close to what one can obtain from solid methane but would solve the problems associated with the use of solid methane in high radiation fields. The concept is based on a heterogeneously cooled pellet bed from which the pellets would be periodically removed to release the stored energy with minimum perturbation of the source operation. In addition to methane other potential moderator materials are examined.

1. Introduction

With new, high power spallation sources currently being proposed and designed in several locations world-wide [1], [2], [3], [4], [5], the need for efficient cold moderators that can operate at source beam power levels of 1 MW and above becomes pressing. Users requests to the moderator characteristics [6], [7], that resulted from extensive consultations are summarized in Table 1.

Table 1:. *Users' requests to moderator characteristics for ESS and SNS to meet the requirements of the next generation pulsed source instruments and science Numbers before and behind the / refer to the high repetition and low repetition rate target stations respectively. ESS planners were not offered an intermediate temperature option*

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

Keywords: Cryogenic moderators; methane; methane hydrate, radiation effects; density of states

This table clearly demonstrates the trend to take advantage of two possibilities in the next generation neutron sources

1. to exploit the high phase space density of neutrons at low energies offered by cryogenic moderators and

2. to use the extra neutron flux on these sources to improve the resolution of the instruments by working with narrow pulses of good shape.

In a workshop sponsored by the OECD and held at Argonne, Ill., in September 1997, possibilities of improving cryogenic moderators for pulsed sources were scrutinized in detail [8] and several opportunities for improved understanding of cold moderators and for novel developments were identified. One outcome of this conference is that the ESS-task group on cold Moderator development that had existed previously was expanded into a world wide collaboration to which representatives of the following institutions have, so far, agreed to subscribe:

Forschungszentrum Juelich (FZJ), Germany) *
Paul Scherrer Institut (PSI), Switzerland *
Risø National Laboratory, (RNL) Denmark *
Rutherford Appleton Laboratory (RAL), UK *
Argonne National Laboratory (ANL), USA
Brookhaven National Laboratory (BNL), USA
Centro Atomico Bariloche (CAB), Argentina
High Energy Accelerator Research Organisation (KEK) Japan
Japan Atomic Energy Research Institute (JAERI) Japan
Joint Institute for Nuclear Research (JINR), Russia
Oak Ridge National Laboratory (ORNL), USA
University of Hokkaido (UH) Japan

* Member of the original ESS-task group

The charter meeting of the expanded collaboration (ACoM-2) was held on Feb. 24 and 25, 1998 at Paul Scherrer Institut, Switzerland. At that meeting the author of this paper was charged to act as spokesman of the collaboration. A Memorandum of Understanding was drafted that was subsequently sent out to the interested laboratories for comments and completion. A follow-up meeting was hosted by Oak Ridge National Laboratory on June 11 and 12, 1998, in which details of the responsibility to be taken on by the participating institutions were worked out. The present paper describes the scientific and technical ideas underlying this collaboration and the working plan the group agreed upon.

2. The potential and characteristics of cryogenic moderators

Cryogenic moderators have become an essential asset in the utilization of research neutron sources over the past decades. This is true for continuous sources such as SINQ and research reactors as well as for pulsed sources. While both types of sources have the common goal of providing the highest possible flux of cold neutrons, pulsed sources have the additional requirement of a short pulse duration. This makes the route chosen for some continuous sources, namely a relatively large volume of 20 to 30 liters of liquid deuterium at 25 K embedded in a thermal moderator vessel impractical. Such cold moderators are of the "re-thermalizing" type, i.e. they accept neutrons in thermal equilibrium at ambient temperature from their surroundings and establish (more or less) a new equilibrium at their low temperature. By contrast, pulsed source cryogenic moderators should slow down the source neutrons as fast as possible and in as small a volume as possible all the way from their original energy of around 1-2 MeV through almost nine orders of magnitude in energy. Design criteria and engineering aspects for cold moderators have been reviewed recently [9] and only a brief summary will be given here:

Clearly, Hydrogen, being of the same atomic mass as a neutron, is most efficient in picking up a large fraction of the neutron's energy in a single collision and is, therefore, the most desirable moderator substance. The cross section of hydrogen and hence the mean free path between two collisions being nearly constant in the whole slowing down regime, the time a neutron lives in a given energy interval is inversely proportional to its velocity. Consequently the product of the neutron velocity and pulse width is a constant whose magnitude is inversely proportional to the number of hydrogen atoms per unit volume. Another important requirement to a good cryogenic moderator material is that it should have sufficiently low-lying modes in its energy spectrum to which slow neutrons can couple. While liquid hydrogen has a rather low density, a material with very good properties in this respect is solid or liquid methane which has nearly free rotational modes at 1 meV. It is for this reason that early, low power pulsed spallation sources used solid methane at low temperatures (around 15 K) with great success. Gains of a factor of 3.5 in cold neutron intensities for wavelengths above 0.4 nm for solid methane relative to liquid hydrogen have been measured for an identical geometry for the time average flux [10], with even better performance in the pulse. Gains may be somewhat lower if the moderator geometries are optimized individually, but will still be more than a factor of 2.5, which certainly makes a solid methane moderator a highly desirable feature on any pulsed neutron source. However, even at a few kW of beam power (ESS is planned to run at 5 MW !), a phenomenon called "burping" has been observed, which manifested itself in a sudden energy and gas release after a certain irradiation dose and which can lead to the destruction of the container. This phenomenon now is largely understood and solid methane moderators are still in use at low power sources but must be warmed up at regular intervals to release the stored energy. In view of these difficulties, intermediate power sources (ISIS, 160 kW) use liquid hydrogen as their cryogenic moderator and liquid methane at 100 K. However, even liquid methane suffers from radiation damage and the radicals form higher polymers that cling to the walls of the moderator vessel and tubes and eventually lead to complete blockage of the system. Currently ISIS has developed a system that allows online replacement of the liquid methane every two days (i.e. after 0.3 MWd), but still the moderator vessel is replaced twice a year (i.e. after 300 mAh or 10 MWd) because the polymerization products continue to build up [11].

3. The general concept of an advanced cryogenic moderator

Since solid, hydrogen rich materials with low-lying energy modes clearly offer the best chance to accomplish the goal of an improved moderator system, ways should be sought to use a good solid moderator. While the need for heat removal, together with the low thermal conductivity at low temperatures, requires the use of relatively finely dispersed material, the desire to achieve a fairly high average hydrogen density let the collaboration choose a concept, in which pellets - preferably spherical ones- would be cooled by liquid or supercritical hydrogen and would be removed from the moderator vessel after as short enough residence time to avoid spontaneous release of stored energy. According to first estimates this would have to happen every 20 minutes, or so [12]. One possibility to accomplish this was presented at the ICANS XIII-meeting [13]. It involves the use of a continuously operating mechanical device (e.g. a helical screw bar) to remove the moderator pellets from the vessel. In contrast to the original concept which foresaw melting of the pellets after each passage, the preferred solution now is periodic heating to a temperature where the stored energy is released (Fig. 1). This makes the overall process technology somewhat easier. Batch disposal of the pellets would become necessary when the performance of the moderator starts to degrade due to non-annealable radiation effects.

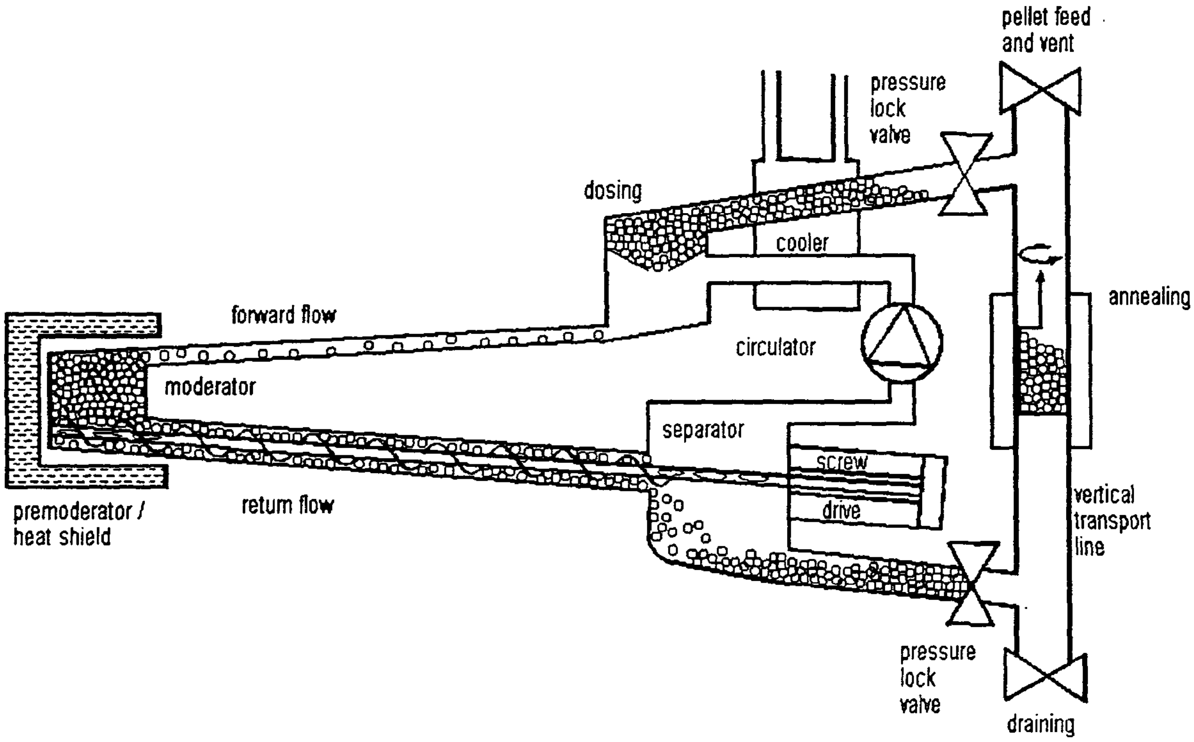

Figure 1: Scheme of a pelletized moderator system with mechanical removal of the pellets from the moderator vessel and recirculation after heating for stored energy release.

A second possibility to transport pellets into and out of the moderator vessel that is more in line with current concepts of moderators that are supplied from and removed to the top of the target block is also considered [14]. This involves fluidizing the pebble bed prior to emptying and "blowing" the pebbles out pneumatically by reversing and increasing the flow of hydrogen. This concept requires some extra volume in the moderator vessel to enable fluidization. The consequences of this "void" and other technical requirements on neutronic performance will have to be studied in detail.

Other options, such as in situ melting of the pellets are also considered, but it will have to be shown that this does not defeat the idea of removing the polymerization products without giving them a chance to cling to the walls and build up a solid or tar-like layer there. This option also depends strongly on the type of moderator material chosen.

4. Some candidate moderator materials

The most popular material for cryogenic solid moderators is clearly methane, which is known to have worked well in low power applications due to the nearly free rotational states of the CH_4-molecule at low temperatures, which gives rise to energy levels at 1 meV and above. Although a wealth of studies on the lattice and molecular dynamics of methane have been performed, little is known on its physical and mechanical properties. Collecting such information is important if transport by mechanical or pneumatic means is foreseen, because excessive deformation, abrasion or breaking of pellets would certainly be difficult to cope with.

Another opportunity to take advantage of the nearly free rotation of methane in a cage is the use of methane hydrate (clathrate). Under certain conditions significant amounts of methane can be stored in the cage-structure of ice. Ice is known to exist in a large number of different modifications, depending on the environmental conditions. The structure that can store methane (and other "small" molecules) is shown in Fig. 2. The degree of filling of the cages depends on pressure and temperature (Fig. 3) but, when all possible cages are occupied, up to 0.17 CH_4-molecules per H_2O-molecule (y=1) can be stored, increasing the hydrogen density by as much as 34 %. From the phase diagram of the CH_4-H_2O system, shown in Fig. 4, it can be seen that making clathrate, while not excessively difficult, is not as straight forward a process as making ice. Suitable methods to fabricate clathrate pellets will have to be developed by the collaboration, if this system is to be used. Fortunately, once trapped, the methane escapes from the ice only very slowly when kept at atmospheric pressure around -20°C.

Experimental data on the frequency spectrum of methane-hydrate are shown in Fig. 5. It can be seen that, compared to the frequency spectrum of water (which is well reproduced in the top frame, cf. Fig 6), the low energy modes are very intense. Note the change in scale between the frames.

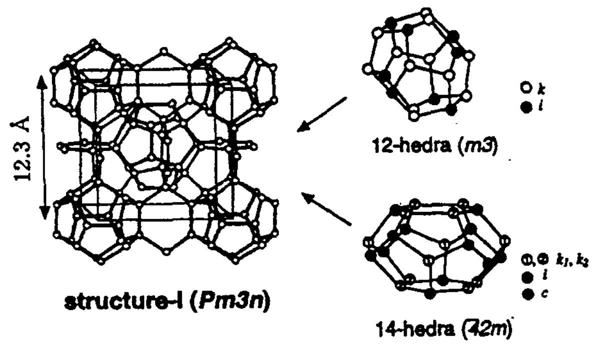

Figure 2: Structure of Hydrates (Structure 1), composed of 12-hedral and 14-hedral "cages" in which methane molecules can be "trapped".

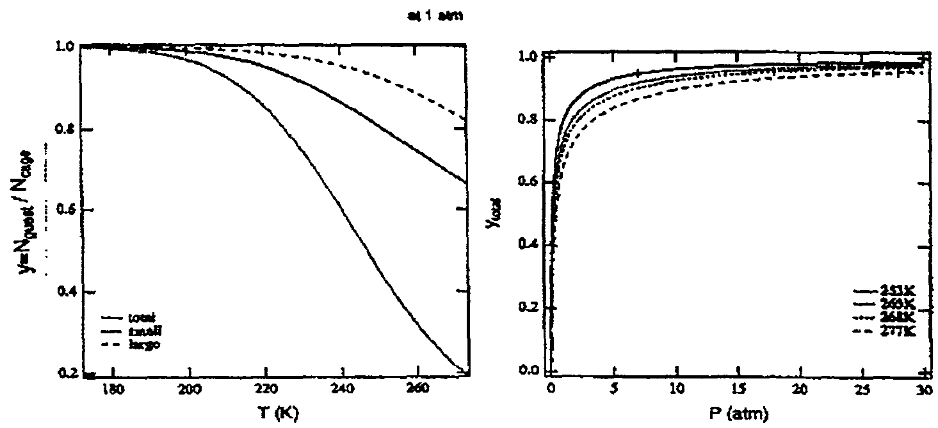

Figure 3: Degree of occupancy of the ice cages by methane as a function of pressure and temperature; y=1 corresponds to ~0.17 CH_4-molecules per H_2O-molecule

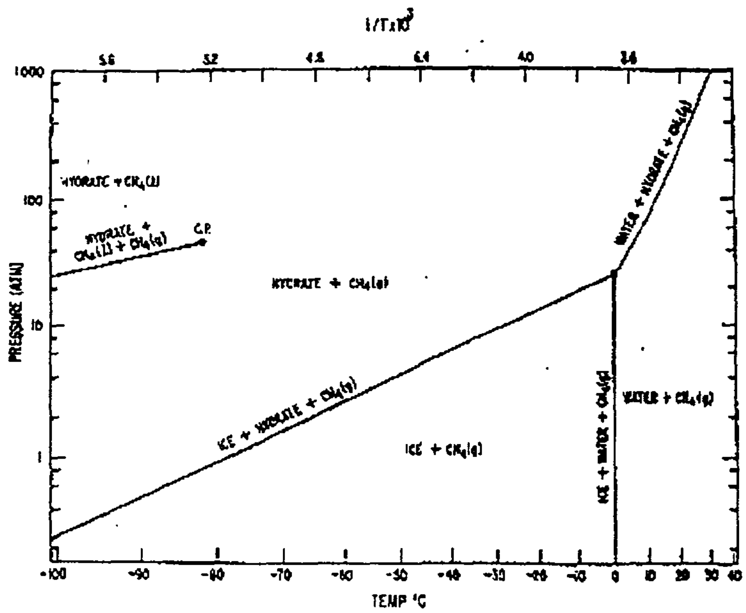

Figure 4: Phase diagram of methane-water

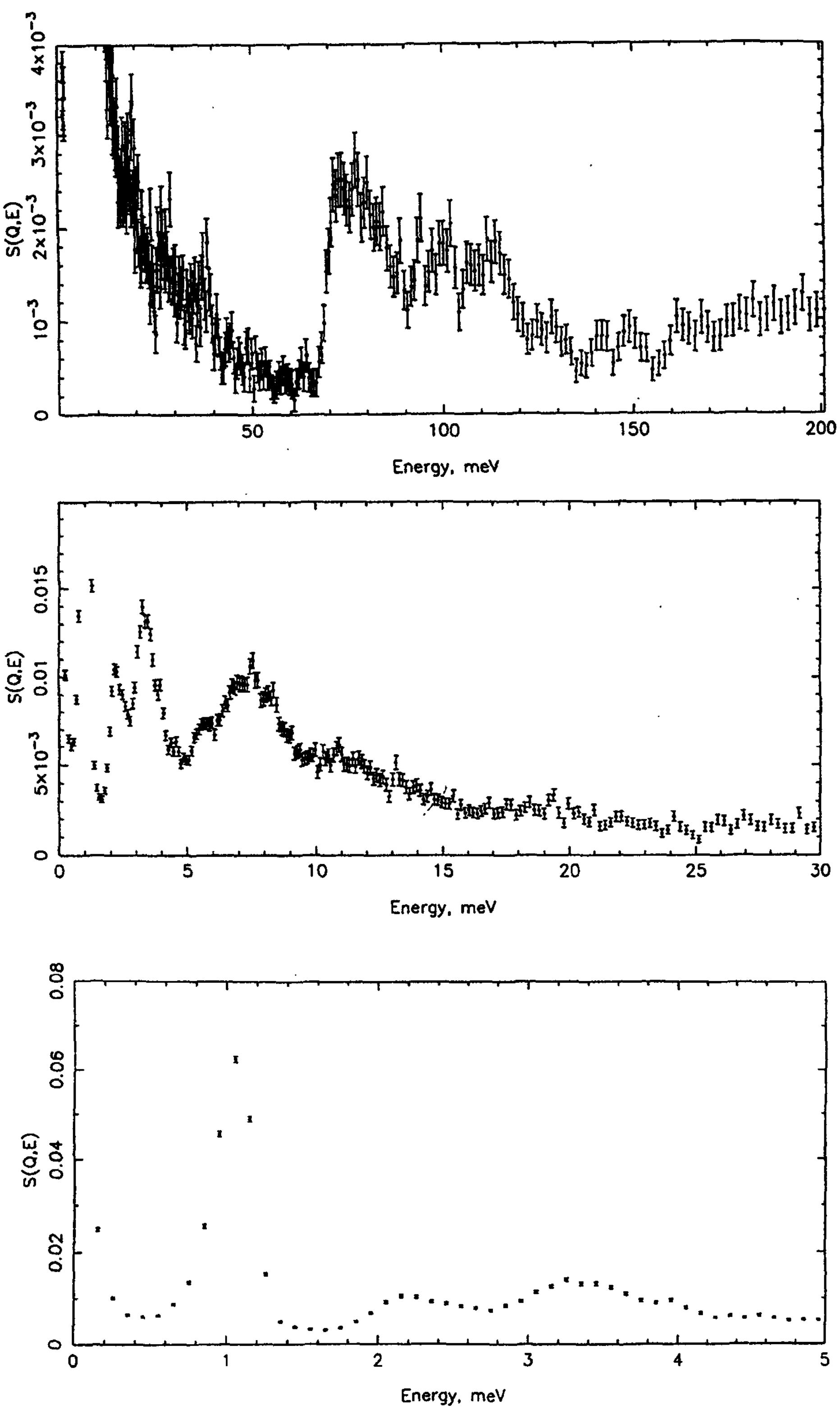

Figure 5: Frequency spectrum of methane hydrate [15]. The three frames show consecutively enlarged sections of the low energy spectrum. Note the change in scale from top to bottom.

The respective spectrum of pure water ice is shown in Fig. 6. In contrast to methane hydrate, pure water ice has no significant density of states below 7 meV and most of the spectrum is concentrated above 65 meV. (This part is also clearly seen in the top frame of Fig. 5).

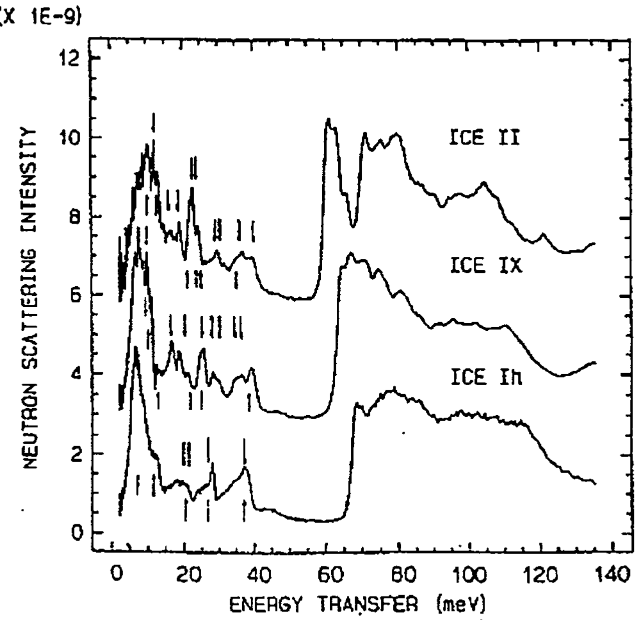

Figure 6: Frequency spectrum of H_2O ice, measured by incoherent inelastic neutron scattering (from [16]). The arrows above the spectra refer to peaks seen in the Raman and those below to peaks seen in the IR.

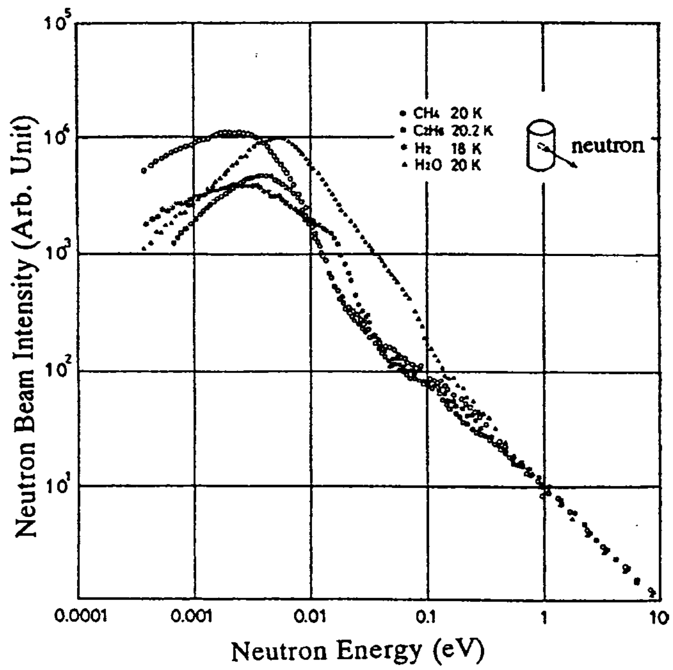

Figure 7: Measured neutron spectra from different moderator materials at 20 K [17]

From measured neutron energy spectra for different kinds of moderators, shown in Fig. 7, it is obvious that H_2O-ice is an excellent moderator down to its lowest frequency mode of about 7 meV. Below 5 meV CH_4 is clearly superior. The hope associated to the use of liquid hydrogen cooled methane hydrate clearly is that the favorable properties of the three materials could be combined to generate a moderator of high performance in the whole energy regime of interest.

Although numerous questions relating to this system, such as radiation damage, mechanical properties at 20 K etc. must still be investigated and several technical problems must be solved, the prospect of improving the performance of cold sources in this way make this a worthwhile effort.

Although methane hydrate, next to methane, is currently the main focus of R+D activities in the collaboration, other opportunities of finding suitable moderator materials are explored as well. This includes examination of different substances, such as propane, and ways to store them in solid materials, in particular different kinds of zeolites or other cage structures.

5. The ACoM working plan

In its charter meeting on Feb. 24 and 25 at Paul Scherrer Institut, Switzerland, the collaboration agreed to set a goal for the decision on the final system to be pursued, which is the end of the first quarter of the year 2000. Until then the following milestones have been agreed upon:

01/1999 Working model(s) for pellet transport at room temperature ready

06/1999 Moderator materials: neutronic performance examined in low power facilities; scattering kernels developed and integrated in NJOY

03/2000 Moderator materials: radiation effects assessed
pellet production: method demonstrated
decision on proceeding with full scale low temperature test rig (outside radiation field)

If funding for the final test rig can be found, the goal would be to have a facility working outside a radiation field by the end of the year 2002. This facility would then be used to gain "hands on" experience with such a system and to study its behavior under various conditions before a final cryogenic moderator for use on a spallation source could be designed.

As a more short term solution to the problem of increasing the moderator performance in the intermediate energy range, the ORNL group is following up on the concept of placing a thin ("semi-transparent") liquid H_2 moderator in front of an ambient temperature water moderator in order to profit from the high phase space density regions of both types. While this is not expected to match an optimized pellet system in the whole spectral region, calculations have yielded promising results and the system would certainly be much simpler and is based exclusively on existing technology. The collaboration as a whole supports this effort.

6. References

[1] "ESS, A Next Generation Neutron Source for Europe"; Vol. III, The ESS Technical Study; G.S. Bauer, T.A. Broome, D. Filges, H. Jones, H. Lengeler, A. Letchford, G. Rees, H. Stechemesser, G. Thomas, eds. The ESS Council, ESS-96-53-M; ISBN 090 237 6659 (1997)

[2] B. Appleton, SNS status report, this conference

[3] M. Furusaka, JHF status report, this conference

[4] T. Mukaiyama, Y. Oyama, M. Mizumoto, T. Takizuka, R. Hino "Neutron Science Project at JAERI" Proc. Top. Meeting on Nuclear Applications of Accelerator Technology, Albuquerque, ANS Order No. 700249 (1997) pp 398-404

[5] G.J. Russell, E.J. Pitcher, P.D. Ferguson, B.W. Patton "Long Pulse Spallation Source Neutronic Performance" J. Neutron Research, **6,** (1997) pp. 33-62

[6] "ESS A Next Generation Neutron Source for Europe", Vol.2, The Scientific Case; ESS Council, (March 1997), ISBN 090 237 6 608

[7] Instrumentation for a High-Power Pulsed Spallation Neutron Source" Proc. of an NSNS workshop held at Oak Ridge, Ten Oct 31 - Nov 1, 1996, ORNL (May 1997)

[8] Proc. International Workshop on Cold Modeators for Pulsed Neutron Sources Argonne, Ill. Sept 28 -Oct 2, 1997

[9] G.S. Bauer "Pulsed Neutron Source Moderator Design and Engineering" in [8]

[10] R.K. Crawford, P. Thiyagarajan, J.E. Eppreson, F. Trouw, R. Kleb, D. Wozinak, D. Leach " The New Small Angle Diffractometer SAND at IPNS" Proc. ICANS XIII; PSI-Proceedings 95-2 (1995) pp 99-117

[11] T.A. Broome, "ISIS Target and Moderator Materials" Proc 2nd Int. Workshop on Spallation Target Materials Technology, Ancona, I; report Jül 3450 (1997) pp55-69

[12] E. Shabalin, this conference

[13] T. Lucas, G.S. Bauer, C.D. Sulfredge "A Pelletized Solid Methane Moderator for a Medium-to-High Power Neutron Source" Proc. ICANS XIII PSI-Proceedings 95-2 (1995) pp 644-653

[14] H. Barnert-Wiemer, this conference

[15] F. Trouw, Argonne National Laboratory; private communication

[16] J.-C. Li, J.D. Londono, K.D. Ross, J.L. Finney, J. Tomkinson, W.F.Sherman, "An inelastic incoherent neutron scattering study of ice II, IX, V and VI in the range frim 2 to 140 meV" J. Chem.Phys. **94** (10) 1991,p 6770

[17] K. Inoeu, H. Iwasa ,Y. Kiyanagi, J. At. Energy Soc. Japan, **21** (1979) 865