# A combined $H_2/CH_4$ cold moderator for a short pulsed neutron source

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## Foreword

Both the ISIS (Rutherford-Appleton Laboratory) spallation source and the Los Alamos Neutron Scattering Center (LANSCE) were designed to produce neutrons as a result of an 800-MeV proton beam being incident on a target. Both systems are intended to accept beam intensities up to 200 μA.

Cryogenic moderators of liquid hydrogen and methane are either in use or are planned for service at both facilities. Very low temperature methane would be an ideal moderating material as it has a high hydrogen density and many-low frequency modes, which facilitate thermalization. Such moderators are in service at two major world facilities, KEK (Japan) and Argonne National Laboratory (USA).

Unfortunately, solid methane has very low thermal conductivity (see Fig. 1) and is subject to radiation damage making a moderator of this type impractical for use in high- intensity beam, such as indicated above. This report outlines a possible alternative using small spheres of solid methane in a matrix of supercritical hydrogen at 25 K.

## General approach

A novel method of building a low-temperature methane moderator might be to use small spheres in a flowing medium of supercritical hydrogen at 25 K. Initial calculations indicate that a diameter of 1.5 mm is about the optimum size. At 200 μA, such a sphere would receive about 1.0 mW of neutron energy. The heat flux from the inner core to the outer surface, assuming a temperature gradient of 30 K, would be about 1.25 mW for an outer surface temperature of 25 K. This would mean a maximum temperature within the sphere of about 55 K, which is 35 K below the freezing point. Unless neutronically unacceptable, this assumption would seem to form a reasonable base for subsequent design calculations. The temperature gradients in present solid moderators are not known, but it is suspected that they might not be too different from the figure mentioned above.

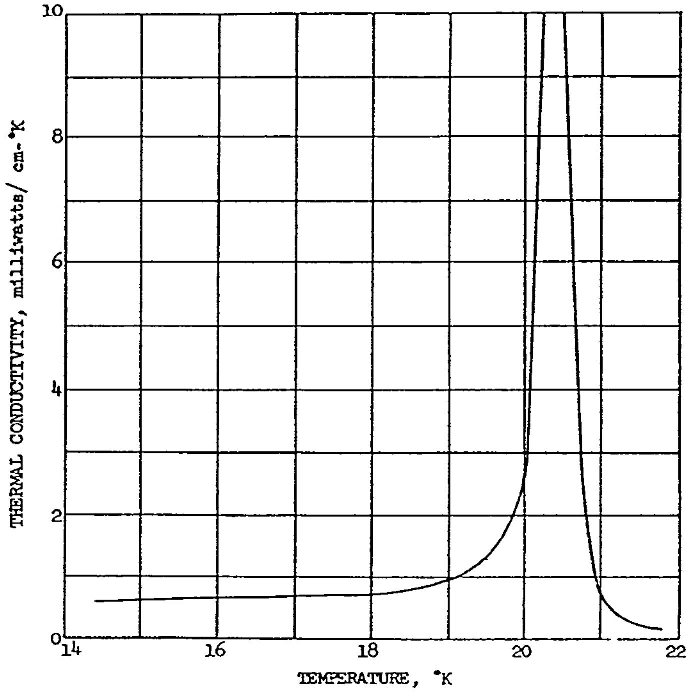
Sources of Data:

Gerritsen, A.N. and van der Star, P., Physica, 9, 503-12 (1942).

Commun. Kamerlingh Onnes Lab. Univ. Leiden, Commun. No. 265c.

Comments:

In the graph below the discontinuity that occurs at the transition temperature (20.4°K) is similar to that found for the specific heat.



KDT/VJJ Issued: 8/28/59

Fig. 1 Thermal conductivity of solid methane (15° to 21°K).

## Proposed system of operation

Uniformly sized spheres can theoretically be packed to a density of better than 73%. However, for practical purposes, an estimated density of 60% has been assumed. The hydrogen density of this mixture would be 0.05 atoms/Å$^3$, which is intermediate between methane and hydrogen (i.e., 0.06 and 0.042 atoms/Å$^3$).

To transport a mixed fluid of this density through pipework into a moderator would be impossible. Therefore, the only recourse is to generate the spheres within the moderator vessel. This would require a cooling region at the top of the vessel outside of the neutron flux large enough to freeze and cool incoming liquid methane. Liquid methane at 95 K would be injected through nozzles in short bursts to form spheres naturally by surface tension. To maintain repeatability would require careful attention to such factors as "duration of burst" and nozzle diameter. Cooling such a sphere to 25 K throughout would require 210 mJ. Because the heat flux is limited to 1.25 mJ/s, the time required in the cooling region would be about three minutes. This figure can now be used to give a methane flow rate and a sphere production rate, which works out to be 250 spheres per second for a moderator volume of one liter. Each sphere, assuming uniformity of flow, would then remain in the moderating region for about 20 minutes.

To assist a steady downward movement of the spheres and prevent block-ups, a vibrator would be attached to the moderator vessel. Such devices are driven by gas and operate by the oscillation of a steel ball in a circular cell. They are available commercially, though not specifically for use at cryogenic temperatures; this should not present a problem if stainless steel is used throughout.

The above sphere-production rate represents a liquid flow of 30 $cm^3$/min, which is too low for a continuous flow system. It would be better, therefore, to maintain a liquid-methane flow of ~150 $cm^3$/s and pass the majority through a bypass restrictor. A gas-operated cryovalve (see Fig. 2) would admit liquid to the nozzles as required, and a second cryovalve in the outlet methane pipe would be used during the initial start up of the system as described later. Pulsations in the flow of the methane would be provided by a motor-driven oscillator connected into the flow line. Together with the liquid-methane circulator, this would produce pressure cycles ranging from zero to double the steady-state pressure produced by the circulator.

All equipment at the moderator would obviously have to be radiation hard making the use of inorganic materials essential.

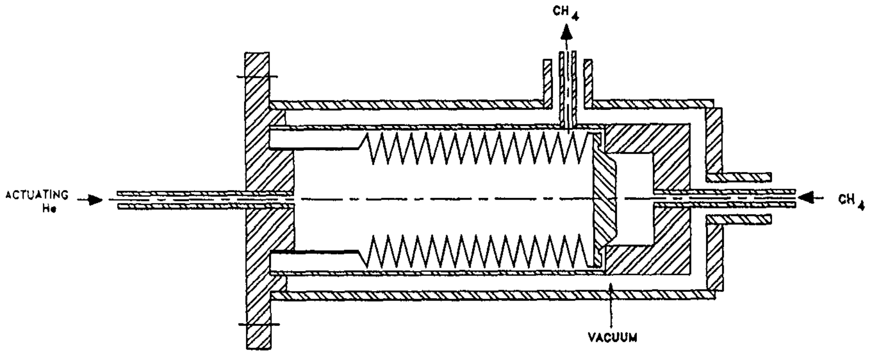


**Fig. 2** Remotely operable cryo-valve.

## Construction

The moderator vessel would constitute a vertical chamber with outward curving sides on account of the high operating pressure of 15 bar absolute. An additional height of ~25 mm outside the neutron beam would rob the target of reflector material. Likewise, a liquification region at the bottom would represent a further loss of ~12 mm in length. Unfortunately, this is the price to be paid for such a system and it must be balanced against the neutronic gains to be made.

Hydrogen would enter at the top of the vessel and exit through the end of the central vertical tube terminating just above the bottom of the moderating region. A strainer across the open tube would prevent the entry of methane particles. Methane would also enter at the top through a series of nozzles. Fully cooled spheres would then progress down the vessel, be melted off in a pool at the bottom, then drawn off into the return pipe. The flow-rate of hydrogen would probably be around 500 cm$^3$/s, although this could be increased to improve the skin coefficient of heat transfer of the sphere surfaces if necessary. A heating block to raise the sphere temperature above 90 K would be attached to the bottom of the vessel using warmed helium gas in a closed loop. Because the vessel wall would have a temperature gradient of 65 K, a thermal break in the form of a local thinning of the wall would be machined into the vessel. Also, the vessel would be tapered outwards by ~3 mm over its height to assist the steady fall of the spheres (see Fig. 3).

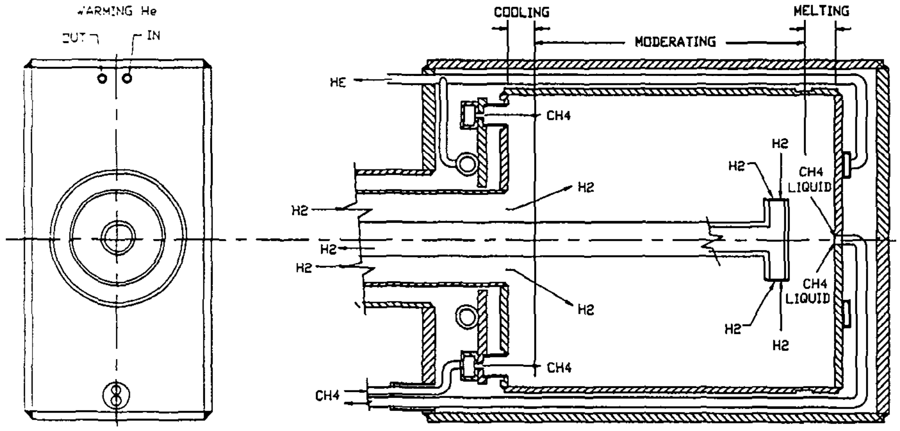


**Fig. 3** Moderator canister.

## Refrigeration

The hydrogen refrigerator would be called upon to freeze and cool the methane spheres and also to remove nuclear heating during moderation. The calculated total load at 25 K would be about 510 W. The methane heat load would only represent transport losses plus losses incurred during purification, i.e., 200 W at 95 K. This heat load would probably be removed by the same refrigerator by tapping into the circuit at an earlier stage (see Fig. 4).

## Installation and operation

The complete system would involve a complex of several feed and return loops. In addition to the hydrogen and methane systems, there would also be helium gas supplies to the warming block, the cryogenic valves, and the vibrator unit.

The most difficult operation would be that of initial start up. The two cryogenic methane valves would remain shut, though the methane circuit could be cooled

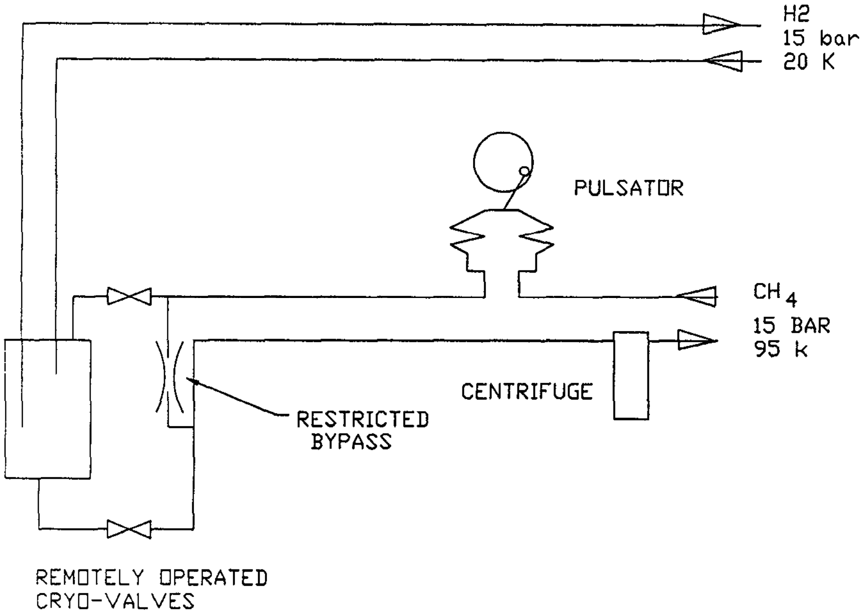


**Fig. 4** Flow diagram.

through its bypass, while the full compliment of hydrogen at 15 bar absolute and 25 K was generated. The methane inlet valve would now be opened with the pulsator operating and the spheres formed. A careful check on the methane gas added together with a check on the hydrogen displaced would enable a good evaluation to be made of the progress. When "full", the exit/return methane valve would be opened. Confirmation of the existence of a pool of liquid at the bottom would be provided by a thermocouple able to differentiate liquid from gas; or, perhaps a platinum thermometer mounted vertically could actually measure the depth of liquid. Once the system was operational, the inlet valve would be opened and closed automatically in response to the liquid methane pressure or the liquid-level indicator. Under normal operation, liquid would prevent any hydrogen entering the methane exit, but during start up, any hydrogen penetrating the methane would be expelled as gas (at 95 K at the refrigerator end of the system).

## Development

The design of such a system could only be entertained with the backup of considerable experimentation. Initial work on sphere formation could be readily carried out using water in liquid nitrogen. This would ultimately have its limitations as the spheres would have a density of about 0.9 gms/cm$^3$ in a fluid of 0.8 gms/cm$^3$ density. The density of solid methane is 0.522 gms/cm$^3$ and that of supercritical hydrogen only 0.0669 gms/cm$^3$. Further work could be done using water in helium gas at 25 K and 10 bar absolute with a density of 0.025 gms/cm$^3$. Finally, methane in helium would give a close approximation of the true fluid state.

## Conclusion

The overall challenge would be of a high order, but what calculations have been done indicate that thermodynamically such a system is feasible. If realized, such a system would provide significant improvements in the shape of the neutron pulse that is desired for pulsed source diffractometers.