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Cold Moderators for Pulsed Neutron Sources

John M. Carpenter
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Argonne National Laboratory

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John M. Carpenter
Argonne National Laboratory
Argonne, IL 60439

Abstract

This paper reviews cold moderators in pulsed neutron sources and provides details of the performance of different cold moderator materials and configurations. Analytical forms are presented which describe wavelength spectra and emission time distributions. Several types of cooling arrangements used in pulsed source moderators are described. Choices of materials are surveyed. I examine some of the radiation damage effects in cold moderators, including the phenomenon of "burping" in irradiated cold solid methane.

I. Introduction

As is the case in steady source (reactor) cold moderators, the main purpose of cryogenic moderators in pulsed sources is to produce long wavelength neutrons for neutron scattering and other applications. In pulsed sources there are additional strong motives for use of cold moderators. They provide short pulses at intermediate wavelengths where ambient temperature moderators exhibit a long tail associated with the Maxwellian spectrum. Even at very short wavelengths, the pulses can be made narrower in some cryogenic materials than in ambient temperature materials due to their significant thermal contraction and consequent higher proton densities at low temperature. Furthermore, some materials with attractive thermalization properties condense and can be used only at low temperatures.

In pulsed sources nuclear heating caused by gamma rays, fast neutrons and slow neutrons is substantially less than in reactor sources because the time average fluxes are a few orders of magnitude smaller, at least in present day pulsed sources. Furthermore pulsed source moderators are typically smaller than reactor cold sources; together with the lower heating rates this implies that

they require substantially smaller refrigerators than do reactor cold moderators. Radiation damage rates in pulsed sources are also lower than in reactors, which leaves more opportunity for the use of hydrocarbons as moderators. Because of the small volume of pulsed source moderators the inventory of combustible material and the related hazards are smaller than in the larger, D_2 systems in reactors.

Pulsed sources present a more detailed set of requirements and a greater range of requirements for different applications for cold moderators than do steady sources. But because of their lower time average power there are a greater range of prospects for meeting these requirements and for optimizing moderators for different applications than there are in reactors.

Figure 1 shows a typical arrangement of a target, moderator and reflector in a pulsed neutron source. To make the most of the available neutrons from the primary source, moderators are very close to the target. A reflector of Beryllium, for example, or other material surrounds the target and moderator to minimize leakage of neutrons from the moderator during slowing down, and to give neutrons which otherwise would have missed the moderator a chance to collide and slow down there. A layer of slow-neutron absorbing material (decoupler) surrounds the moderator and the neutron beam path to prevent long lived neutrons from the reflector from entering the moderator and broadening the pulse. In many cases a sheet of slow-neutron absorbing material (heterogeneous poisoning) is included below the viewed surface of pulsed moderators to control the width of the pulses.

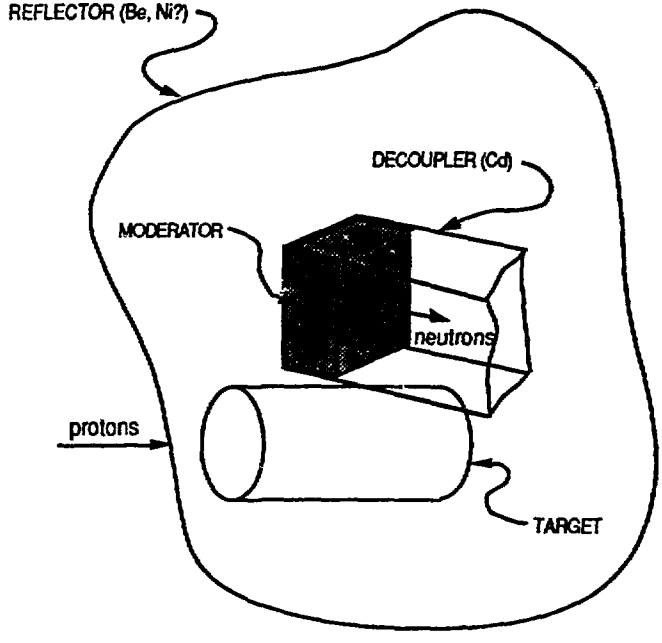

Figure 1. A typical arrangement of a target, moderator and reflector in a pulsed spallation neutron source. Targets are around 20 cm in length and 10 cm in diameter; moderators dimensions are about 5x10x10 cm.

In the sections below, I review the installations of cold moderators in pulsed sources.and present some analytical functions which describe the integrated intensities and the emission time distributions as functions of wavelength. Some arrangements used for cooling cold moderators in pulsed sources are described. Choices of moderating materials are surveyed. I examine some of

the radiation damage effects in cold moderator material, including the phenomenon of "burping" in cold solid methane.

II. Currently Operating Pulsed Sources and Cold Moderators

There are four pulsed spallation neutron sources and two pulsed reactors now operating for slow neutron research. Table I lists the pulsed spallation sources. All these have at least one cryogenic moderator. One of the early Argonne prototype spallation neutron sources, ZING-P' (operated from 1977 until 1980) had a liquid Hydrogen moderator. Two pulsed reactors IBR-30 and IBR-2 are operating at Dubna; plans are afoot to install a cold moderator in the 2 MW IBR-2.

TABLE I

Currently Operating Pulsed Spallation Neutron Sources

Source	Location	Target	Proton Current, μA	Proton Energy, MeV
IPNS	Argonne	235-enriched U	16.	450.
ISIS	Rutherford	238-U, Ta	110.	750.
KENS	KEK (Tsukuba)	238-U	10.	500.
LANSCE	Los Alamos	Tungsten	70.	800.

Until 1988, the IPNS target was of depleted Uranium. From time to time ISIS has used a Tantalum target. Originally the target of ZING-P' was of Tungsten; later the first depleted Uranium target was tested in ZING-P'.

Table II lists the cold sources now in operation or which operated in the past at pulsed spallation sources.

TABLE II

Cold Moderators at Pulsed Spallation Neutron Sources

Source	Designation, beams	Material	Temperature (K)	Dimensions (cm)	Poisoning
IPNS	"C" 1,2,2',3	L-H_2	20.	10.x10.x7.6	none, grooved
IPNS	"F" 1-3,4-6	L-CH_4	110.	10.x10.x5.	0.5 mm Cd @ 1.7 cm
IPNS	"H" 1,2,3	L-CH_4	110.	10.x10.x5.	0.5 mm Cd @ 2.5 cm.
ISIS	N 4,5,6	L-H_2	35.	11.x12.x8.	none
ISIS	N 1-3, S 6-9	L-CH_4	100.	11.5x12.x4.5	none
KENS		S-CH_4	25.	12.x15.x5.	none
LANSCE		L-H_2	20.	12.x12.x7.	none
ZING-P'	"A"	L-H_2	20.	10.x7.6x5.	none

The L-H_2 moderator at ISIS is actually supercritical Hydrogen at about 15 atm pressure.

The small dimensions of pulsed source moderators reflect the compromise of narrow pulse widths against time average intensity which is appropriate for traditional time-of-flight applications--the pulse widths at low neutron energies are principally determined by leakage. The small size of the moderators is matched to the small size of the primary neutron source, that is, the target. Furthermore, the typical dimension of the moderators is comparable to the mean free path of primary neutrons from the target and to the migration length of fast neutrons in the material.

Calculations of the performance of pulsed sources and moderators almost inescapably have to be performed with 3-D Monte Carlo codes with generalized geometry capability. This is because of the presence of strong absorbers, because the neutron spectrum varies greatly from region to region in the target-moderator-reflector assembly and because of the complexity and asymmetry that is typical of the spatial arrangements of moderators, beam paths and targets in pulsed sources. Further, it is necessary to recognize that the neutron population is driven by an external source; reactor criticality codes may need to be adapted for this application.

Accelerators, targets, moderators and neutron scattering instruments for spallation sources are the subjects of meetings of the International Collaboration on Advanced Neutron Sources (ICANS), held every other year. The most recently published are the proceedings of the 1988 meeting[1].

III. Materials for Pulsed Source Moderators

Protons are most efficient for slowing down fast neutrons because their mass is the same as that of the neutron and because their low energy neutron scattering cross section is larger than that of any other practical species. Except for considerations of finer detail addressed below, materials with high proton densities are preferred for pulsed source moderators. Table III lists a number of materials which are or may be useful as cryogenic moderators in pulsed sources.

TABLE III

Pulsed Source Moderator Materials

Material	Temperature, K	Proton Density, protons/Å3	Melting and Boiling (1 atm) Temperatures, K
H_2O (for reference)	293	0.067	273, 373
$(CH_2)_n$, 0.94 gm/cm^3 (ref)	293	0.081	-------
TiH_2 (for reference)	293	0.095	-------
H_2	20	0.042	14, 21
CH_4	109	0.070	90, 110
CH_4	10	0.079	90, 110
C_2H_6	165	0.068	90, 184
C_3H_8	228	0.064	83, 231
$C_{12}H_{18}$, Mellitine	293	0.047	439, 538
C_9H_{12}, Mesitylene	293	0.039	228, 438

Water (ice), polyethylene and metal hydrides in and of themselves are not effective cryogenic moderators materials because they have too few low frequency modes to provide efficiently for the final stages of thermalization, although their proton densities are quite high. Liquid Hydrogen is quite good in this regard, but the large (15 meV) rotational level spacing makes for large differences in the scattering between the low temperature para form and the normal ortho-para mixture; its low proton density leads to broad pulses of short wavelength neutrons. Liquid and solid methane are very attractive because of their good low-temperature thermalizing power, and have high proton densities as well. Ethane and propane are similarly attractive, with even lower rotational level spacings than methane, but have fewer rotational modes per proton than methane. Mellitine and mesitylene have freely rotating methyl groups but even fewer low frequency modes per proton and lower proton densities. They are attractive because they remain in condensed form at room temperature.

IV. Cooling Systems

The cooling systems of cryogenic moderators in pulsed sources are typically smaller than those in reactors, having a capacity of a few tens or hundreds of watts because of the smaller heat loads. Figure 2 illustrates three types, the recirculating liquid system, the recirculating liquid-gas system and the stationary system.

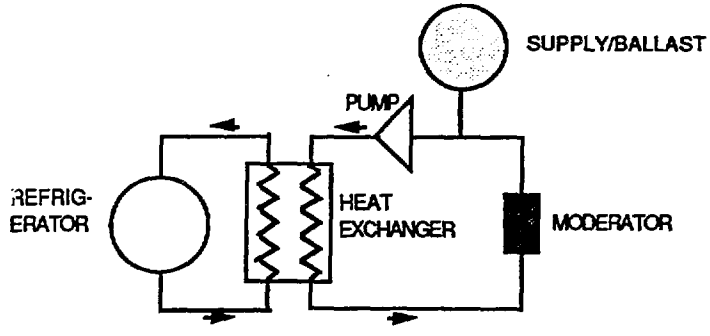

Figure 2a. Recirculating liquid system.

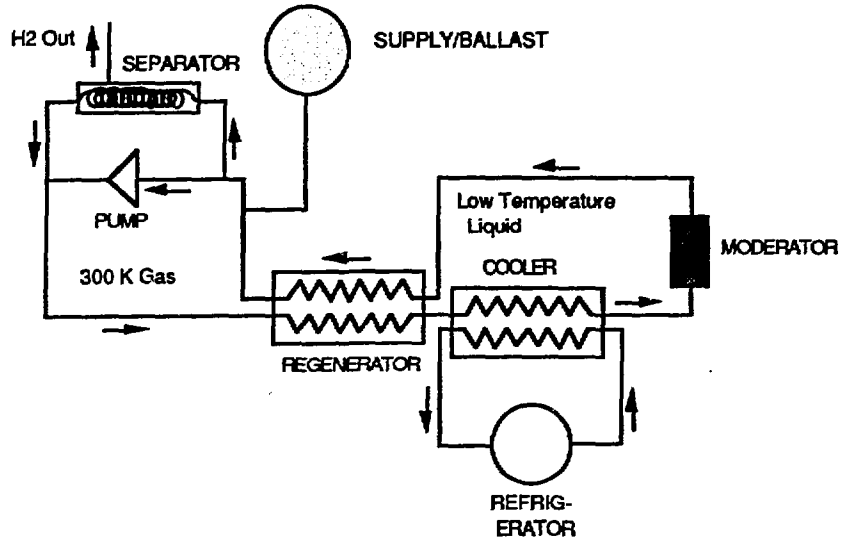

Figure 2b. Recirculating liquid-gas system.

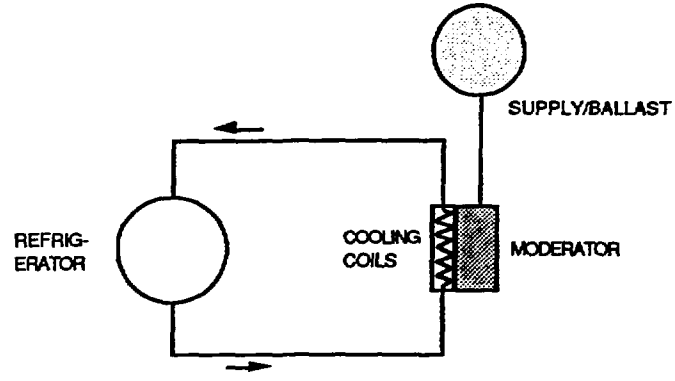

Figure 2c. Stationary moderator system.

The "C" moderator in IPNS is a stationary system which can be filled with either Hydrogen or methane; cooling is provided by a 300 watt closed cycle Helium liquefier. The "F" and "H" moderators in IPNS are circulating liquid-gas systems; cooling is provided by evaporating liquid Nitrogen, through an intermediate loop of Helium gas. Both the liquid Hydrogen and the liquid methane moderators in ISIS are circulating liquid systems; cooling is provided by closed cycle Helium refrigerators. The liquid Hydrogen moderator in LANSCE is a circulating liquid system, also cooled by a closed cycle Helium refrigerator. The solid methane moderator in KENS is a stationary system, cooled by a Displex closed cycle Helium refrigerator. The liquid Hydrogen moderator in ZING-P' was a naturally circulating thermosyphon system.

The low thermal conductivity of hydrogenous solids at cryogenic temperatures requires that measures be taken to transport heat from within the moderators. Figure 3 illustrates two ways in which this has been accomplished.

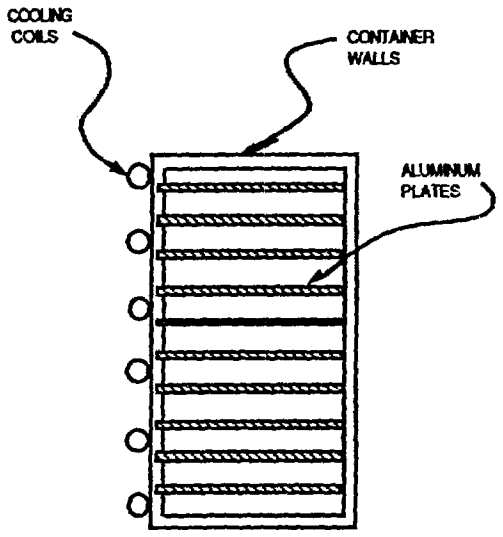

Figure 3a. The KENS internal cooling system for solid methane.

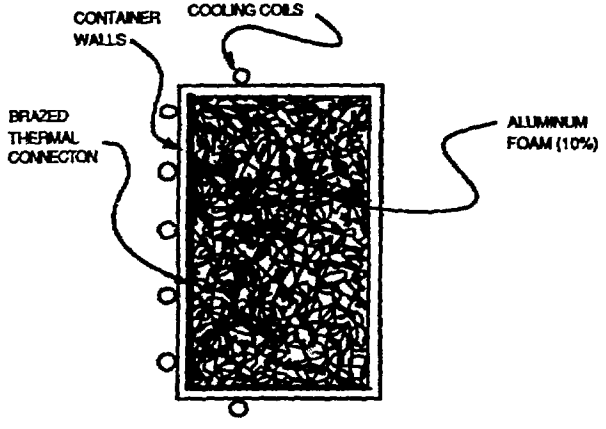

Figure 3b. The IPNS internal cooling system for solid methane and liquid Hydrogen.

V. Radiation Damage

Aside from its eventual appearance as sensible heat, nuclear radiation, neutrons and gamma rays, produces chemical changes in cold moderator materials. In some cases this is harmless, but in other cases this leads to accumulation of troublesome products, and in solid methane it leads to temperature instabilities and damaging pressure excursions.

Hydrogen

Fast neutron and gamma ray interactions in Hydrogen H_2 and D_2 produce recoiling free protons and deuterons,

$$H_2 \xrightarrow{n_f,\ \gamma} H + H \qquad (a)$$

Recoiling protons can induce the same reaction as they slow down. In both the liquid and the gas the products harmlessly and rapidly recombine due to the fast diffusion of the ions

$$H + H \longrightarrow H_2 + \text{heat} \qquad (b)$$

I do not know how rapidly this recombination takes place in the low temperature solid.

Hydrocarbons (e.g. CH_4)

In methane and other hydrocarbons the principal initiating radiolysis reaction is (for example in methane)

$$CH_4 \xrightarrow{n_f,\ \gamma} CH_3 + H \qquad (c)$$

which is rapidly followed by recombination according to

$$H + H \longrightarrow H_2 + \text{heat} \qquad (b)$$

from which H_2 accumulates and also according to

$$H + CH_4 \longrightarrow CH_3 + H_2 + \text{heat} \qquad (d)$$

which takes place rapidly and produces more CH_3, and

$$CH_3 + CH_3 \longrightarrow C_2H_6 + \text{heat} \qquad (e)$$

In liquid (and gas) the recombination according to (e) takes place quickly due to the rapid diffusion of methyl radicals. In the solid the diffusion rate can be very slow, leading to long-term storage of chemical energy which can be released upon externally induced warming or externally controlled changes in the coupling of the cooled material to the coolant, or released spontaneously after building up to critical concentrations. This is the origin of the "burping" phenomenon which is discussed further below.

Recombination according to (e) is not the only possible result. It has been known for a long time that in gaseous methane a single initiating event can lead to recombination into quite heavy hydrocarbons, up to about 20 carbons long, albeit with lower probability than (e). We have observed this in liquid methane, where the heavier products build up more-or-less in proportion to the time or accumulated dose. This is evidence for the direct formation of heavier hydrocarbons as opposed to a sequential buildup which would lead to t^n behavior of the concentration of, say, C_nH_x. Although the intermediate weight hydrocarbons are volatile and soluble in methane and have thermophysical properties rather like methane (see Table II), the heavy hydrocarbons can rapidly exceed solubility limits and collect in pipes and elsewhere to obstruct flows and inhibit heat transfer. Table IV shows the results of gas chromatographic analysis of the contents of the circulating loop of the IPNS "F" moderator system, after one week's operation starting with a

fresh charge of pure methane. The accelerator delivered an average current of 15 μA to the Booster Target; the H_2 separator was not in operation.

TABLE IV.

Molar Concentrations of Compounds in Gas from the Circulating Side of the IPNS "F" Moderator After One Week of Operation at 15 μA with the Booster Target

Material	Concentration
H_2	22.6 %
CH_4	75.2 %
C_2H_4	< 0.16 %
C_2H_6	1.50 %
C_3H_8	0.39 %
C_4H_{10}	< 0.07 %

The distribution of radiolysis products seems to be different in solid methane than it is in liquid methane, based on analyses of the volatile products of irradiation in liquid and solid moderators in IPNS.

H_2 can accumulate beyond the solubility limits both in the liquid and in the solid material. In circulating liquid methane systems the Hydrogen-rich vapor phase, even at the level of a few mole percent H_2, can accumulate to exclude a significant fraction of the liquid phase, due to the low density of the vapor. In solid methane excess H_2 collects but has a low vapor pressure at temperatures below about 20 K. When the temperature rises, Hydrogen trapped in the solid exerts its higher vapor pressure on the entire solid and its container. If the container is insufficiently compliant and the compressibility of the solid is insufficient to accommodate the expansion of trapped Hydrogen, the resulting stresses can cause the container to fail. This has happened in IPNS, once in a single event which led to very high pressures, and in other instances where numerous stress cycles led to fatigue failures.

Water

In water (H_2O or D_2O) radiolysis leads to formation of H_2 (D_2), OH radicals, O_2, O_3 and possibly H_2O_2 as stable or metastable products. At low temperatures energy can be stored in the form of these reactive species as in cold solid methane. Even though the only eventual stable product of recombination is again water (unless some product is removed), the potential for release of stored energy and for pressure increases due to expansion of trapped gases is similar to that in solid methane.

Degradation of Performance

In addition to causing operational problems, radiation damage effects of various kinds accumulate to degrade the neutronic performance of most materials, largely because of the loss of bound protons and partly because of the formation of Hydrogen and heavy hydrocarbons. This is most important in the case of solid moderators, such as polyethylene and solid methane[2].

VI. Wavelength Spectra

Of the two principal measures of the performance of pulsed source cold moderators, spectra and pulse shapes, the wavelength spectra are easiest to measure. Figure 4 shows the usual arrangement.

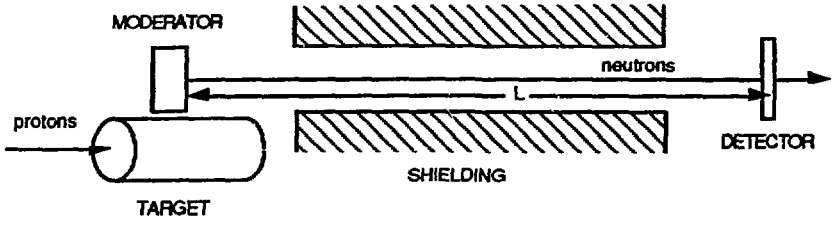

Figure 4. Arrangement for measuring the wavelength spectrum of a pulsed source moderator.

The time average counting rate in the detector $C(t)$ per unit time-of-flight t is

$$C(t) = A\eta(\lambda)\varphi(\lambda)(h/mL) + B \qquad (1)$$

where B is the background (primarily due to delayed neutrons from fissionable targets), A is the area of the beam incident on the detector, $\eta(\lambda)$ is the detector efficiency for neutrons of wavelength λ, $\varphi(\lambda)$ is the time average flux per unit wavelength at the location of the detector, (h/m) = 3955.4 Å-m/sec is the ratio of Planck's constant to the neutron mass, L is the distance from the moderator surface to the mean position of the detector and (ignoring wavelength dependent emission time delays and shifts of the mean detection position)

$$t = (h/mL)\lambda\,. \qquad (2)$$

In this context it is conventional to express fluxes as functions of the energy E,

$$E = (\hbar^2/2m)/\lambda^2 \qquad (3)$$

where in practical units $(\hbar^2/2m) = 81.787$ meV-Å^2. The flux per unit energy $\phi(E)$ is related to the flux per unit wavelength

$$\phi(E) = (\lambda/2E)\varphi(\lambda) \qquad (4)$$

The flux is related to the "beam current" of the moderator $i(E)$ (in analogy with light sources, $i(E)$ is a luminous intensity, equal to the total number of neutrons per unit time per unit energy, per steradian in the direction of the beam)

$$\phi(E) = i(E)/L^2 \qquad (5)$$

which is an intrinsic property of the moderator. When the detector is a low efficiency "1/v" detector,

$$\eta(\lambda) = K\lambda, \tag{6}$$

where K is a constant, and the counting rate is proportional to $E\phi(E)$

$$C(t) = 2AK(h/mL)E\phi(E) \;\alpha\; E\phi(E). \tag{7}$$

Ignoring corrections due to the attenuation of neutrons by air or structural material in the beam (or assuming that these corrections have been made) the flux can usually be accurately represented

$$\phi(E) = \phi_{MB}(E) + \phi_{SD}(E). \tag{8}$$

except in the important case of liquid hydrogen moderators. $\phi_{MB}(E)$ is a Maxwellian function

$$\phi_{MB}(E) = (E/E_T^2)e^{-E/E_T}\,\phi_{Th}, \tag{9}$$

with mean energy (the "thermal energy")

$$E_T = k_B T_{eff}. \tag{10}$$

$k_B = 0.086165$ meV/K is Boltzmann's constant and T_{eff} is the "effective temperature" of the distribution, which is always greater than the physical temperature of the moderator. $\phi_{MB}(E)$ is normalized

$$\int_0^\infty \phi_{MB}(E)dE = \phi_{Th}\,, \tag{11}$$

where ϕ_{Th} is the time average "thermal" neutron flux. In the case of small moderators, especially liquid Hydrogen moderators, the low energy flux distribution is not of Maxwellian form. (It is beyond the scope of this paper to discuss the origin of this phenomenon and some of the general features of the time and wavelength distributions which are the basis of the present discussion. Underlying is the extensive theory of time-dependent neutron thermalization. The reader interested in further exploration may profitably consult, for example, the excellent treatise of Williams[3] or the illuminating discourse by Parks, Nelkin, Beyster and Wikner[4].)

$\phi_{SD}(E)$ is a "slowing-down" function

$$\phi_{SD}(E) = (1/E)(E/E_{Ref})^{\alpha}\Delta_1(E)\;\Delta_2(E)\;\phi_{epi}\,, \tag{12}$$

where ϕ_{epi} is the time average epithermal neutron flux, a constant given by

$$\Delta_1(E_{Ref})\,\Delta_2(E_{Ref})\,\phi_{epi} = E_{Ref}\,\phi_{SD}(E_{Ref})\,. \qquad (13)$$

E_{Ref} is an arbitrarily chosen reference energy, conventionally 1. eV, and α is the "leakage exponent, a positive number usually less than 0.1.

$\Delta_1(E)$ is the "joining function". Several empirical forms have been used to represent $\Delta_1(E)$; we use a generalization of Westcott's joining function,

$$\Delta_1(E) = \frac{1}{1 + (E_{co}/E)^s}\,, \qquad (14)$$

where s is the "cutoff exponent" and E_{co} is the "cutoff energy"; usually around $E_{co} \approx 5.0\ E_T$. To avoid an infrared catastrophe, $s > 2 - \alpha$, but usually $s \approx 5.0$.

In methane it is necessary to account for a prominent bump in the spectrum presumably associated with the high frequency vibrational modes of the molecule. We have characterized this bump entirely empirically in the form of a stretched Gaussian function

$$\Delta_2(E) = 1 + Ce^{-(E^p - E_B^p)^2/2\sigma^2} \qquad (15)$$

where E_B is the energy at which the bump appears, p is the stretching exponent which we find is around -2/3 but do not refine in least squares analysis, and σ is the width parameter. The coefficient C represents the amplitude of the bump.

Usually, $2. < \phi_{Th}/\phi_{epi} < 5.$, a much smaller ratio than is characteristic of steady reactor sources, therefore pulsed sources provide useful fluxes of neutrons at wavelengths far shorter than the peak of the Maxwellian. For most pulsed sources, the time-average epithermal neutron flux is around $\phi_{epi} \approx (3.\text{x}10^{-4}$ neutrons/ster/source neutron) x S/L^2 where S is the time average global leakage rate of neutrons from the primary source.

Nonlinear least-squares fitting algorithms usually converge rapidly and robustly to a well-fitting solution, given a sufficient range of time-of-flight data.

Figure 5 shows spectra for three different moderators. Each curve represents a function fitted to spectra corrected for attenuation in air and Aluminum, then recalculated with the corrections reapplied. Delayed neutron backgrounds are not shown, but represent about 2.8 % (with the Booster Target of IPNS, 0.44 % with the depleted Uranium target) of the total number, spread uniformly across the 1/30 th second long time interval between pulses in IPNS. The relative magnitude of this background thus depends on the distance from the moderator to the detector in which the spectra are measured, but the fraction of delayed neutrons in the background is invariant.

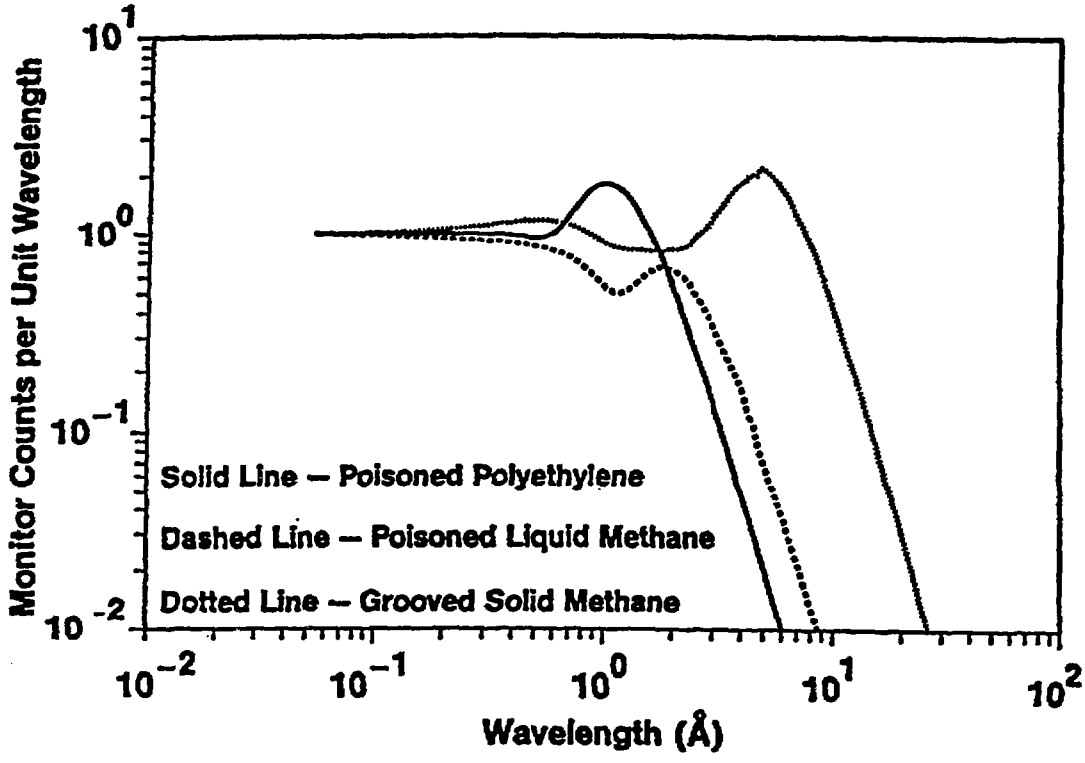

Figure 5. Comparison of flux spectra for 350 K polyethylene, 105 K liquid methane and 20 K grooved, solid methane.

Three spectra are shown, one for ambient temperature (about 350 K) polyethylene poisoned with .5 mm Cd 25 mm below the viewed surface, one for a 105 K liquid methane moderator similarly poisoned 17 mm below the surface and one for an unpoisoned, grooved, 20 K solid methane moderator. Comparing the polyethylene and liquid methane spectra in the range of the Maxwellian peak, two effects are visible: the peak of the Maxwellian is shifted in wavelength as expected according to the square root of the temperature ratio; the more severe poisoning of the liquid methane moderator leads to a significant loss of the integrated intensity in the Maxwellian, even so the flux of long wavelength neutrons from the liquid methane is about 3 times higher. Comparing the solid methane with the polyethylene reveals two effects: the 20 K moderator shifts the Maxwellian to still longer wavelengths, and the grooved, unpoisoned moderator produces a larger Maxwellian flux relative to the epithermal flux, with the result that in the limit of long wavelengths, neutron flux from the grooved, cold solid methane is more than 300 times greater than that from the room temperature polyethylene.

The discontinuity at 4.67Å arises due to the [111] Bragg edge in the scattering cross section of Aluminum and is one of several such features caused by Aluminum windows and other structure in the beam; these serve as markers of the wavelength scale.

Reference (5) contains a survey of the effects of poisoning, cooling and materials choices upon the spectra and pulse shapes.

VII. Pulse Shapes (Emission Time Distributions)

For conventional time-of-flight applications, the width -- indeed the detailed shape -- of the pulse as a function of time is a defining factor in the resolution of instruments. In principle, the shape of the pulse of primary neutrons from the source broadens the pulse from a moderator, but since in all the operating pulsed spallation sources the primary source pulse width is quite short, this broadening is ignored in the present discussion. The pulse shapes are then entirely determined by the features of the moderators, and the pulse shape is a strong function of the wavelength. Reflectors surrounding the moderators also affect the pulse shapes, but effective decoupling reduces these effects to insignificance for neutrons with wavelengths in the range of interest and they are ignored here. It is useful to discuss pulse shapes in two regions, the short wavelength, epithermal range, and the longer-wavelength, thermal range.

Measurements of the Pulse Shape

The pulse of neurons emitted from the moderator is conveniently characterized by the time and wavelength distribution at the viewed surface. Measurements can be made remotely, using a "time- focussed" single crystal diffraction method[6,7], shown in Figure 6. The monochromating crystal is cut perpendicular to the [220] planes; the [111] planes are the reflecting planes.

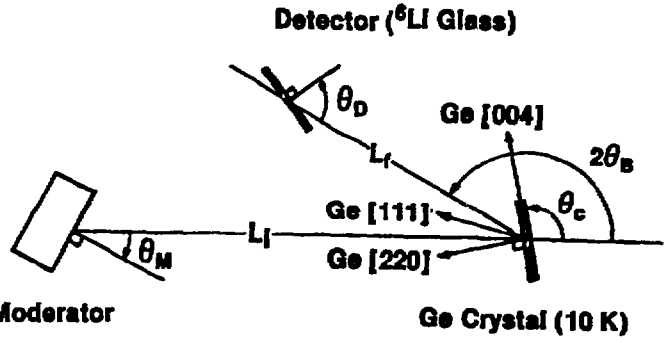

Figure 6. Time Focussed Crystal Spectrometer arrangement for measuring the emission time distribution as a function of wavelength

With such an arrangement, using a thin crystal and a thin detector and having the proper relationships between the ratio of flight paths, moderator viewing angle, crystal cut and detector orientation angles and Bragg angle, the emission time distribution can be measured with negligible time broadening, even when viewing large areas of moderator, crystal and detector. All allowed orders of the aligned reflection are seen, which enables separate determinations of the pulse shape for many wavelengths in a single setup. Cooling the crystal to low temperature enhances the intensity for short wavelengths by reducing the Debye-Waller exponent for high order reflections.

Pulse Shapes for Short Wavelengths.

For short wavelength neutrons, the shape of the pulses is closely approximated by the function describing slowing down in infinite hydrogenous moderators,

$$i(t,\lambda) = (a(\lambda)t)^2 \, e^{-a(\lambda)t} \, \bar{i}(\lambda)/2a(\lambda)f, \qquad (16)$$

where $\bar{i}(\lambda)$ is the time average beam current per unit wavelength, f is the source pulsing frequency and

$$a(\lambda) = v\Sigma_s(\lambda) \qquad (17)$$

in which v is the neutron speed and $\Sigma_s(\lambda)$ is the macroscopic proton scattering cross section. The pulse FWHM is $3.394/a(\lambda)$ and the pulse standard deviation is $\sqrt{3}/a(\lambda)$, and is therefore inversely proportional to the proton density. For short wavelengths, the cross section is constant, so the pulse width is proportional to the wavelength.

For longer-wavelength neutrons, in the range of the Maxwellian, the pulse shape is dominated by the exponential decay of the fundamental eigenfunction of the neutron distribution in the moderator. The pulse has a sharp rising edge which reflects partly the rise of the slowing down function, and partly the convolution of that function with an abruptly turned-on falling exponential. Susumu Ikeda [7] found that the function represented schematically in Figure 7 fits room temperature polyethylene pulse shapes very well.

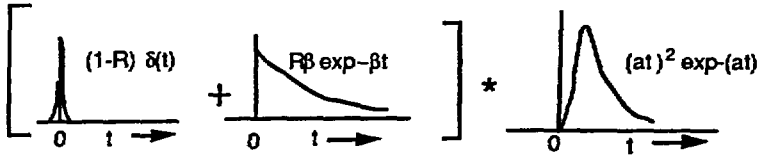

Figure 7. Ikeda's function.

The asterisk denotes convolution. $\delta(t)$ is the delta function, β is a constant, $R(\lambda)$ is the wavelength-dependent fraction of the intensity in the exponentially-decaying "storage" term and (1 - $R(\lambda)$) is the fraction in the slowing-down term. This function does not fit the pulse shapes for liquid methane as accurately as it does for those of polyethylene--a third term seems to be required in this form The form fitted to data for unpoisoned liquid methane by Taylor and Robinson [8], shown in Figure 8,

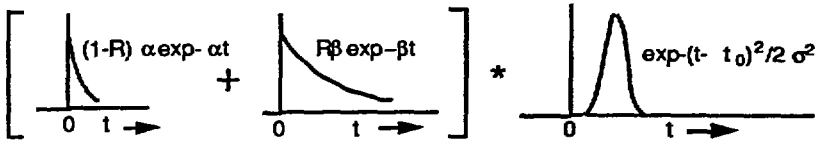

Figure 8. The function of Taylor and Robinson.

fits the data reasonably well. Here, α, t_0 and σ as well as R depend on the wavelength; β is a constant smaller than α. More work needs to be done in characterizing these distributions. Figure 9 shows the shapes of pulses from a poisoned liquid methane moderator and from a poisoned ambient temperature polyethylene moderator, for 2.843 Å and 1.137 Å neutrons. The polyethylene had degraded after lengthy irradiation, and the liquid methane was somewhat less dense than theoretical because of frothing which arose because of the accumulation of Hydrogen vapor in the container.

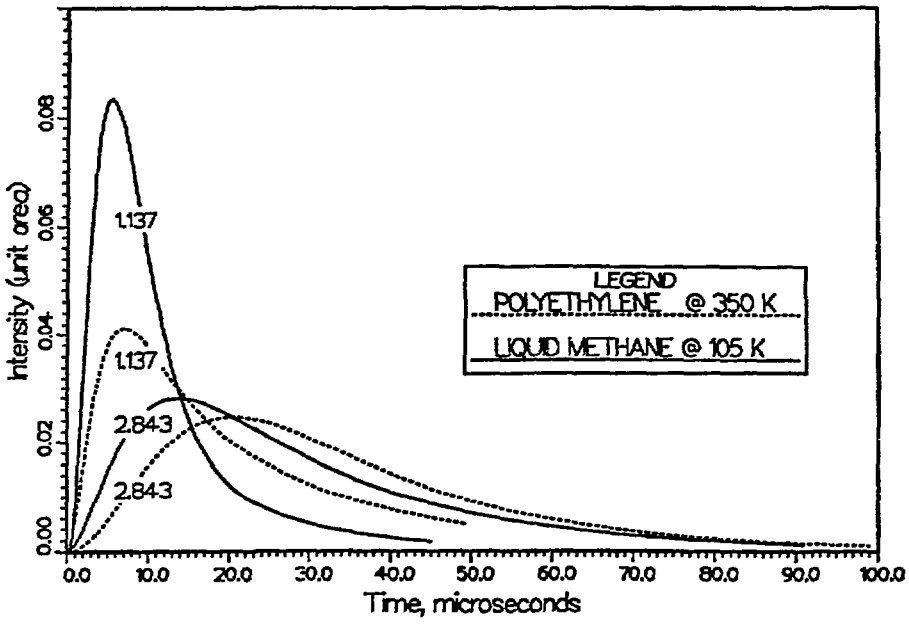

Figure 9. Pulse shapes at two wavelengths for an ambient temperature polyethylene moderator poisoned with .5 mm Cd, 22 mm beneath the viewed surface, and for a liquid methane moderator, poisoned 17 mm beneath the viewed surface.

The poisoning, which is at different depths in the two moderators, mainly affects the length of the decaying tail on the pulses. The pulse width and the integrated intensity of the pulse are adjustable by changing the poison depth. The pulse of 1.137 Å neutrons from the liquid methane is a sample from the epithermal distribution; it does not exhibit much of the slowly decaying exponential, which still influences the pulse of 1.137 Å neutrons from the ambient temperature moderator. The remarkable feature of these distributions is the sharpness of the leading edge of the pulse, which leads to a gain in information in terms of spectrometer resolution. Neutrons thermalize faster in methane than in polyethylene, evidently because of the presence in methane of the rich distribution of low energy modes of excitation,. This produces a much sharper leading edge on the pulses of low energy neutrons from methane than from polyethylene. Figure 10 shows the full width at half maximum of the pulses from the same two moderators, as functions of wavelength. The FWHM is a crude measure of the pulse shapes and does not reveal their details, in particular the sharp leading edges, but the figure shows the general trend of the pulse widths.

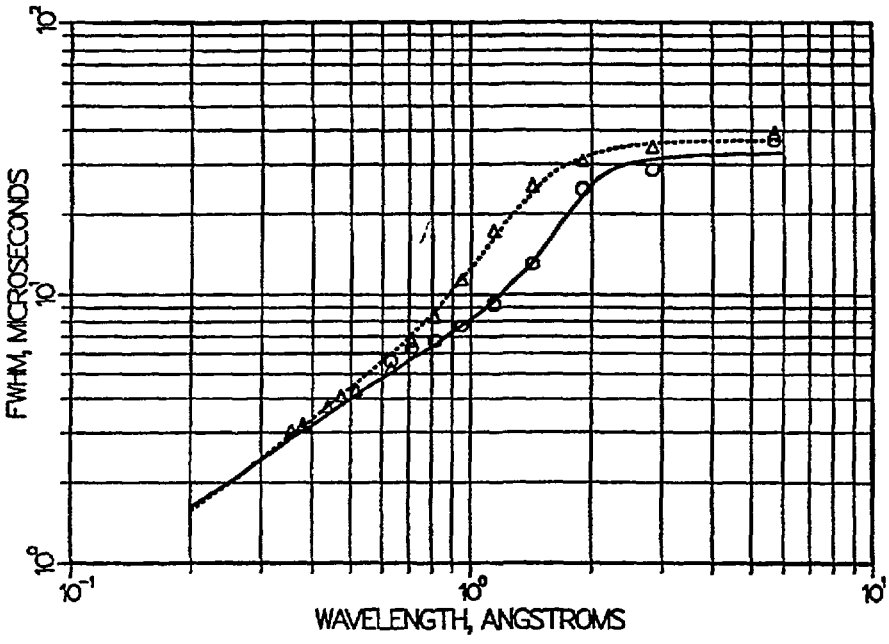

Figure 10. The Full Width at Half Maximum of pulses from a poisoned ambient temperature polyethylene moderator and from a poisoned liquid methane moderator, vs wavelength. The curves are physically-motivated rational polynomials fitted to the data.

At the shortest wavelengths, the pulse widths are proportional to the wavelength. At longer wavelengths, the pulse widths rise, reflecting the presence of the exponentially-decaying fundamental eigenfunction. At longest wavelengths, the pulse widths level off since the decay time of the fundamental mode is independent of wavelength. In the cold moderator, the desirable proportional region extends to longer wavelengths than in the ambient temperature moderator because the Maxwellian spectrum is confined to longer wavelengths. Best resolution in time-of-flight, as measured by FWHM, occurs where FWHM/λ is smallest; this happens at the longest wavelengths.

VIII. Thermalization Properties of Cold Moderator Materials

Eligible materials for cold moderators must exhibit a rich spectrum of low frequency modes which can absorb small quanta of energy, to enable neutrons to equilibrate at low temperature. All materials have low frequency acoustic wave modes or their equivalent, but only the low frequency tail of the Debye spectrum (in solids) or a small component of diffusive scattering (in liquids) is active at low temperatures and at low neutron energies. The best materials for pulsed source applications, in which the moderator is small, have additional low frequency molecular rotational modes, as for example in H_2 and CH_4. Figure 11 shows the rotational level diagrams for H_2 and CH_4.

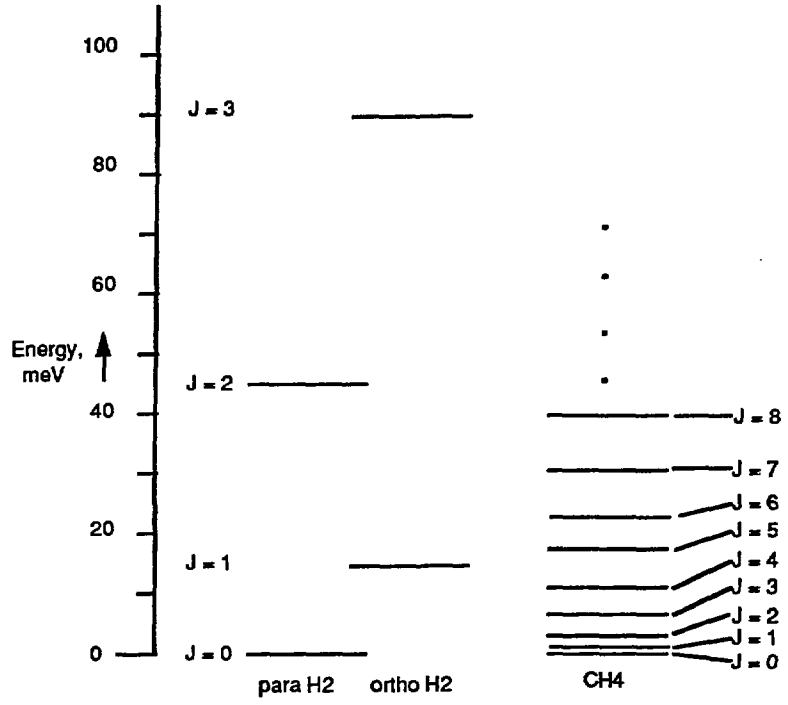

Figure 11. Rotational energy level diagram for hydrogen and methane.

Para Hydrogen, with even angular momentum quantum numbers has the lowest energy and at low temperatures is the predominant phase. However ortho Hydrogen is metastable and in the absence of catalysts requires many hours to convert to para Hydrogen. Neutrons induce the transition from para to ortho and can lose the roughly 15 meV energy difference between the $J = 0$ and the $J = 1$ levels; the minimum energy loss in scattering from ortho Hydrogen is 30 meV. Thus neutrons with energies below 15 meV energy cannot excite rotational transitions.

The total neutron scattering cross section is dramatically different for the two species. Figure 12 shows the total scattering cross sections of normal (ortho:para = 3:1) and para Hydrogen as a function of neutron energy.

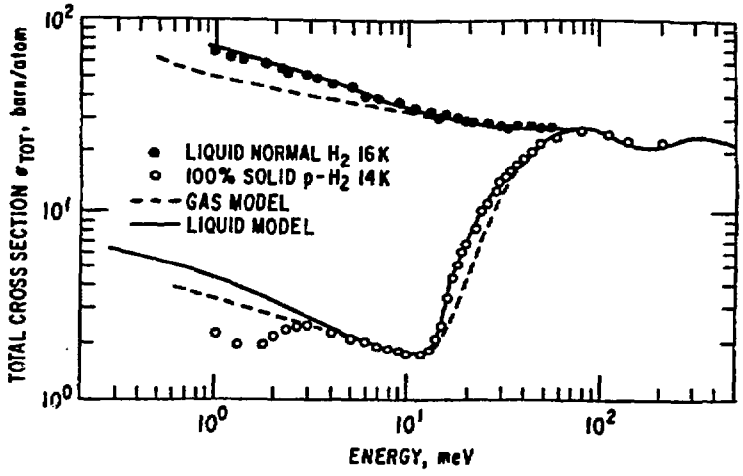

Figure 12. The total scattering cross sections of normal and para Hydrogen.

In ortho (and normal) H_2 the scattering at low energies is elastic insofar as rotational excitations are concerned; only center-of-mass motions can absorb energy in scattering. In small moderators the inferior low energy inelastic scattering properties of para H_2 are compensated somewhat by the low total cross section, which allows low energy neutrons to stream out once they have reached low energies. Figure 13 shows the inelastic scattering function of 9 K solid methane.

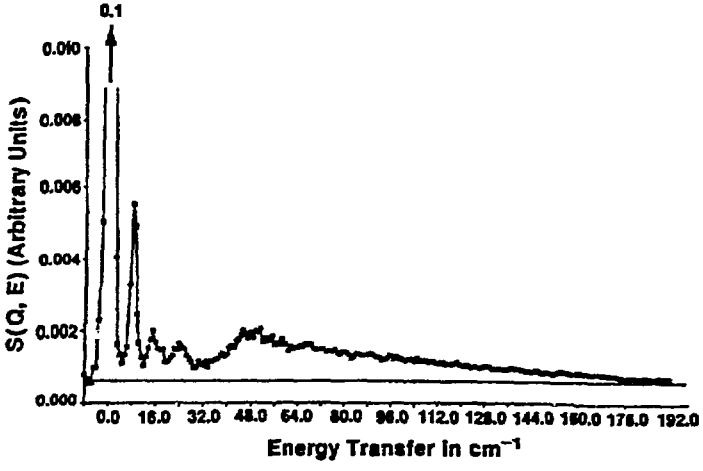

Figure 13. The scattering function of solid methane at 9 K, measured at scattering angle 90° scattering angle and for final energy 3.6 meV. The peaks at 8,16 and 24 cm^{-1} (1., 2., 3. meV) are rotational transitions of the nearly-free methane molecule.

The rotational peaks at 1, 2, 3 meV are responsible for the excellent thermalization properties of solid and liquid methane.

IX. The "Burping" Phenomenon

When cold solid methane is irradiated with high energy neutrons, H_2 and CH_3 accumulate as discussed in section III above. Free protons or monatomic Hydrogen rapidly combines with CH_4 to form H_2 and CH_3. The methyl radicals diffuse (the diffusion is probably a virtual diffusion, wherein the CH_3 identity moves by proton hopping) and recombine only slowly at low temperatures. Then they can accumulate to such densities that upon recombination their heat of recombination raises the temperature of the material significantly, increasing the diffusion rate and triggering further recombination. This happens spontaneously if the rate of production of the methyl radicals ("defects") is large enough, and can also be induced by externally controlled warming or by changing the cooling conditions. The process has been called "burping"[9]. (Identifying methyl groups rather than monatomic Hydrogens as the energy storing defects represents a departure from the discussion in reference (9); this departure is based on more accurate chemical information than I was aware of at the time of writing that paper.) The resulting temperature rise can be sufficient that the vapor pressure of the accompanying trapped Hydrogen reaches several atmospheres, and endangers the container. Figure 14 shows the temperature measured during an induced "burp" in the cold solid methane moderator, which had been irradiated in IPNS for about one day with about 7. x 10^{18} protons on the depleted Uranium target. The energy released, computed from the temperature rise and the heat capacity of the methane and the container, represents about 1.1 joules per gram.

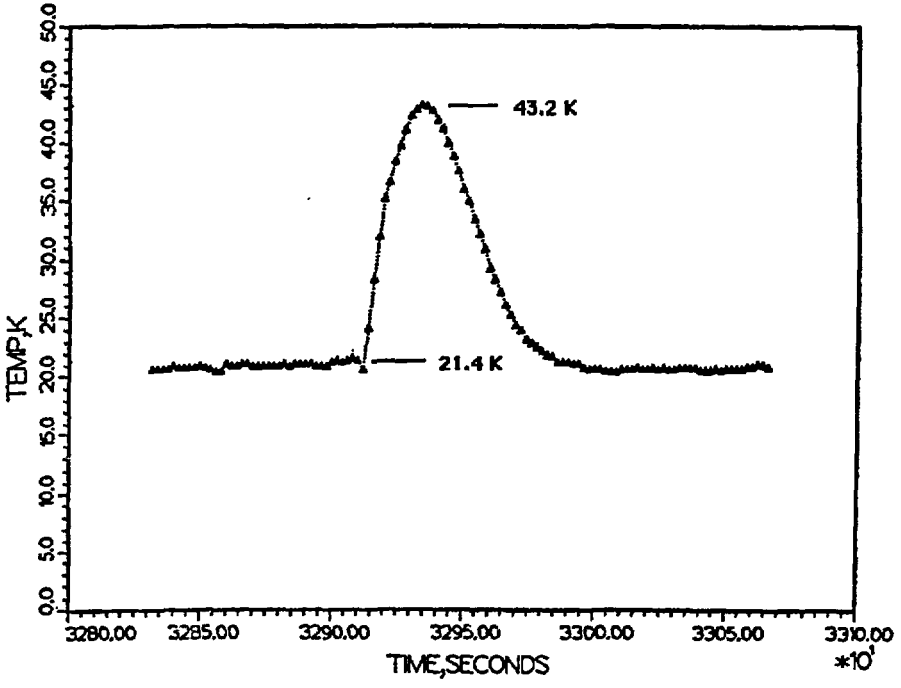

Figure 14. The temperature at the center of the IPNS "C" moderator as a function of time after an externally induced "burp".

A lumped parameter description of the system provides insight into its behavior;

$$\frac{dN}{dt} = R(t) - K(T)N^2 \tag{18}$$

$$MC(T)\frac{dT}{dt} = P(t) + EK(T)N^2 - H(T - T_{cool}(t)), \tag{19}$$

where $N(t)$ is the total number of "defects" (CH_3's), $R(t)$ is the rate of production of defects in the system, $K(T)$ is a recombination rate coefficient, $T(t)$ is the temperature, $C(T)$ is the heat capacity of the system, $P(t)$ is the total power input to the system, E is the energy released per recombination event, $H(T,t)$ is the coefficient for heat transfer between the system and the coolant and $T_{cool}(t)$ is the temperature of the coolant. $C(T)$ and $H(T,t)$ are strong (and unknown) functions of the temperature. (18) assumes binary recombination of defects and (19) assumes Newton's law of cooling. $K(T)$ is assumed to have the Arrhenius form,

$$K(T) = K_0 \exp(-\Delta E/k_B T), \tag{20}$$

where ΔE is an activation energy and k_B is Boltzmann's constant and K_0 is a constant or at worst a mild function of T. $T_{cool}(t)$ and $H(T,t)$ are externally controllable variables. The equations are highly nonlinear, not only because of the binary recombination law and the temperature dependence of the coefficients, but most significantly because of the Arrhenius law dependence of the recombination rate upon the temperature. Of course the system could be more accurately described as a distributed system, but such a description is beyond our current capabilities which are already limited by the lack of detailed information.

Numerical solution of the "burp equations", (18) and (19), reveals that there are two regimes of behavior. When the temperature is high, the defect production rate low, the heat capacity or the heat transfer coefficient large, the system monotonically approaches a state of steady equilibrium. Otherwise, the system exhibits nonlinear, bounded oscillatory behavior which is the "burping" that is observed. Figure (15) shows the result of Runga-Kutta integration (to handle the widely-changing derivatives) of the burp equations for conditions that lead to oscillation; the coefficients are assumed to be independent of temperature, except that the actual heat capacity of the system, including the 20.4 K transition in CH_4, is correctly included. We find that the activation temperature $T_E = \Delta E/k_B$ must be around 150 K.

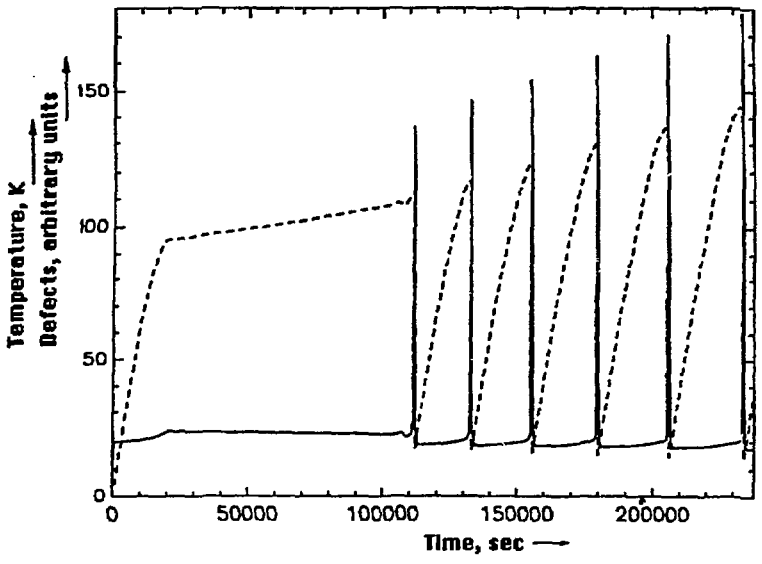

Figure 15. Runga-Kutta integration of the burp equations.

The coolant temperature decreases linearly in the calculation, beginning at a high temperature so that the system does not oscillate. Eventually, the moderator temperature becomes low enough that the system oscillates, beginning with a small ripple just before the large excursion at 110,000 seconds. The frequency of oscillations decreases and the energy released in each excursion increases as the temperature becomes lower. The lumped parameter model seems to be incapable of accurately describing the details of the excursions.

Burping behavior has also been observed in the KENS cold methane moderator, but only since the installation of the depleted Uranium target which increased the radiation damage rate. Presumably that increase pushed the KENS system across the threshold for oscillation. Burping has been observed from the outset of operation of the IPNS cold solid methane moderator. In anticipation of greater burping difficulties upon the installation of the Enriched Uranium Booster Target, we are temporarily using liquid Hydrogen. We believe that it will be possible to live with the burping by following some annealing schedule in which the stored energy is periodically released and, perhaps with lower frequency, the trapped Hydrogen is removed. The KENS moderator is now operated with such an annealing program.

X. Conclusions

Cold moderators using liquid Hydrogen,liquid methane and solid methane are in fruitful use in all the operating pulsed spallation neutron sources. They have been optimized for different applications, but are yet in a stage of rapid development. Problems of clogging and voiding due to the accumulation of radiation damage products remain to be solved even at the present levels of power density and radiation damage density, as do the problems of controlling and accommodating the effects of burping. Methods for characterizing their performance have been reasonably well worked out, and the performance of the existing cold moderators is fairly well understood. Further developments can be expected involving the use of other moderating media and construction material and other modes of operation. Calculations and tests of neutronic performance should be performed to improve the optimization of geometries and to assess new

choices of materials. Further work is called for to improve the operational reliability of existing cold sources, and to provide moderators for sources of higher power in the future.

XI. Acknowledgements

This work was performed under the auspices of the U. S. Department of Energy, Division of Materials Sciences, under Contract W-31-109-Eng-38.

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Responses to Questions on Pulsed Source Moderator Paper

J. M. Carpenter

(P. Egelstaff)

Question: Isn't the burp process the same as the Wigner energy release familiar to operators of nuclear reactors? Can you learn anything from their experience?

Answer: In a very broad sense the processes are similar. However, in graphite, energy is stored in the form of lattice defects while in solid methane energy seems to be stored in the form of reactive methyl radicals. The major difference is that in the methane case the rate of energy release is proportional to the square of the stored energy (methyl radical) density; in graphite the energy release rate is probably a more complicated function of the stored energy density. Both processes are thermally activated but the difference in the density dependence of the energy release rates leads to differences in the mathematical descriptions of the two phenomena and in the details of behavior of the two systems.

In a practical sense, the management of the stored energy may come down to similar procedures, whereby the stored energy is released under control by periodic annealing. Hydrogen produced simultaneously with the methyl radicals by radiolysis exacerbates the problem in solid methane because even modest temperature increases lead to large increases in the pressure of trapped hydrogen.

(R. Pynn)

Question: You mentioned the problem of production of hydrogen in liquid methane moderators. How is this overcome in ISIS?

Answer: I think that the H_2 is exhausted from some high point in the system, say from a surge tank. Some CH4 must be lost with it. It will be best to inquire directly of the ISIS engineers.