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The Development of Solid Methane Neutron Moderators at the Intense Pulsed Neutron Source Facility of Argonne National Laboratory*

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ABSTRACT

The Intense Pulsed Neutron Source (IPNS) started using solid methane moderators in 1985 because of their efficient conversion (about 3.5 times greater than was achieved with a liquid hydrogen moderator) of fast neutrons to long wavelength neutrons. However, the solid methane moderators experienced numerous failures due to pressure surges caused by a combination of (1) the release of stored energy, which occurred when methane radiolytic products recombined, and (2) the expansion of hydrogen, which built up in the solid methane during irradiation. During the ensuing years studies were made to determine how to operate the solid methane moderators without causing failure. The rate at which stored energy built up during irradiation and the temperature at which hydrogen was released during annealing were determined. Since 1993 IPNS has successfully operated the solid methane moderators (at about 30 K) by periodically annealing to the liquid state around 90 K after every roughly three days of irradiation.

1. IPNS OPERATION

1.1 Beam Operation

IPNS is a pulsed spallation source that operates as a national user facility for materials research. The accelerator system produces a 450 MeV proton beam at a time-averaged current of 15 μA. Short (~100 nsec) pulses of protons, at a rate of 30 times per second (30 Hz), strike a depleted uranium (^{238}U) target producing fast neutrons with energies around 1-10 MeV. The fast neutrons are slowed down to energies of 1 eV and below by moderators located above and underneath the target. A graphite and beryllium reflector surrounds the moderators and reflects fast neutrons back into the moderators to enhance the neutron flux. Twelve horizontal neutron beam tubes view selected faces of the moderators and permit neutrons to travel to the neutron scattering instruments. The thermal neutrons are collimated to an appropriate size in each beam line then strike the sample. The neutrons are scattered by the sample and their angle of scatter and energy measured by detectors using time of flight techniques.

1.2 Moderator Operation

IPNS has three moderators (C, H and F) located adjacent to the uranium target. The moderator in the F position is maintained by a closed system of flowing liquid methane. The C and H moderators consist of static solid methane cooled by cold helium gas flowing in channels on the back or sides of the container.

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The solid methane moderators are vacuum insulated containers that have a volume of approximately 600 cc (10 cm x 10 cm x 6 cm). The moderators are first cooled to 110 K by helium gas from a closed cycle helium refrigerator. Methane originally metered into a supply tank then liquefies in the moderator container until 90% full of liquid methane. At this point, filling is stopped and a helium cover gas applied to keep the system above one atmosphere. The moderator is further cooled to an operating temperature of about 30 K.

To prevent problems with the spontaneous release of radiolytically produced energy, the moderator is warmed to the liquid state (90 K) after about 3 days of operation. This annealing of the moderator results in the loss of some methane gas which degrades the neutron intensity over time. Therefore, the moderator gas is changed and fully replenished after approximately 1 week of operation.

2. HISTORY

2.1 Moderator History

IPNS has used many different moderator materials in a search for the most efficient and reliable moderator of neutrons that also provided the neutron spectra and pulse characteristics required for the different types of scattering instruments. Throughout, heterogeneous poisoning was employed to optimize the moderator performance. IPNS began using high density polyethylene at room temperature which deteriorated under irradiation and gave poor resolution for the instruments. The next material tried was cold high density polyethylene which also degenerated under irradiation but provided somewhat better instrument resolution. The next change was to liquid methane at ~100 K which provided a better neutron spectrum, however, "goop" (polymerized product of radiolysis) formed that plugged and coated the heat exchangers. Hydrogen bubbles also were formed from radiolysis causing a loss of density in the liquid methane and subsequent loss of neutron intensity. The hydrogen bubble problem was later resolved by a redesign of the moderator flow pattern. Changing to solid methane[1] provided the neutron intensity and spectrum needed, but was unreliable due to failures caused by radiation induced thermo-chemical instability and hydrogen release. Liquid hydrogen was also used for a time [2], but had ~3.5 times less neutron intensity at long wavelengths than the solid methane.

2.2 Solid Methane History [3, 4]

During the period 1985 - 1988, IPNS operated with a solid methane moderator in the C position. There were numerous failures of the moderator which resulted in a loss of experimental time and costly replacements. The cause was ultimately identified [5, 6] as due to the pressure surge created by the combination of the release of stored energy from recombination of methane radiolytic products and the expansion of hydrogen built up in the solid methane during irradiation. A theory of the effect was formulated which provided guidance in the management of the moderator operation. From October, 1988 until August of 1991, IPNS operated with an enriched uranium (^{235}U) target which increased intensity of the neutron beam ~ 2.5 times and correspondingly increased the nuclear heating and radiation damage rates in the moderators. Realizing that there were already problems with solid methane in the C moderator with the depleted uranium target, IPNS decided to use a liquid hydrogen moderator for the C position and continue with liquid methane moderators for the H and F positions.

After the enriched uranium target expired, IPNS returned to a depleted uranium target with the resultant loss of intensity. To regain the roughly 3.5 times greater intensity at long wave length compared to what liquid hydrogen achieved, the decision was made to return to a solid methane

moderator in the C position, despite the history of many solid methane moderator failures. A moderator improvement committee was formed and scientists at other facilities were consulted for ideas on how to operate the solid methane moderator to avoid failures.

3. OPERATIONAL DATA

3.1 Moderator Experiments

In December of 1992, tests were begun on solid methane moderators in the C position to determine the rate at which the stored energy built up during operation (as a function of both irradiation time and operating temperature) and at what temperature the hydrogen was released during an anneal. Trial operating schedules were determined by observing the stored energy release during recombination as shown by the rapid temperature rise when warming the moderator during an anneal. Figure 1 shows this energy release which increases with the irradiation time between anneals and with the lowering of moderator operating temperature. In addition, spontaneous temperature rises (burps) were observed to take place somewhat irregularly in the C moderator. Such spontaneous burps do not remove all the stored energy. One spontaneous burp of the C moderator occurred after about 3 days of irradiation with a 15 μA proton beam current on the depleted uranium target. The subsequent scheduled anneal produced another release of stored energy following a relatively short irradiation of 1 hour.

The tests also showed that very little hydrogen gas evolved during warming of the solid methane until 65 K was reached. Hydrogen gas comes off rapidly as warming takes place through this temperature up to about 90 K where the methane melts. Figure 2 shows the hydrogen release as a function of annealing temperature as measured with a residual gas analyzer (RGA).

Tests were continued until March of 1993, resulting in the successful operation of the moderator at approximately 30 K when annealed to the melting point of methane, 90 K, every 2-3 days.

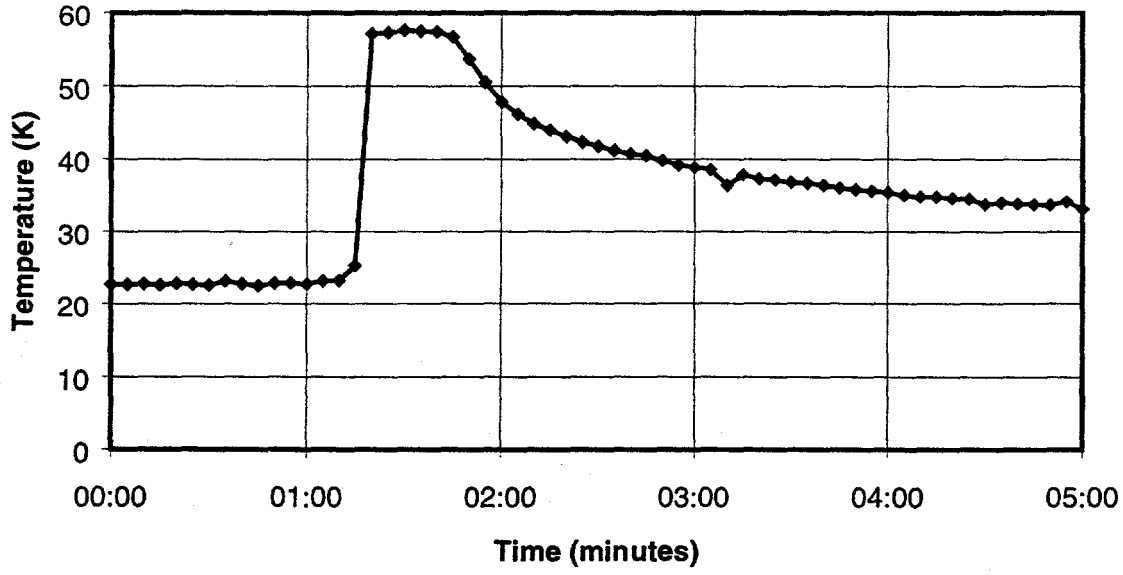

Figure 1. Stored energy release during moderator anneal.

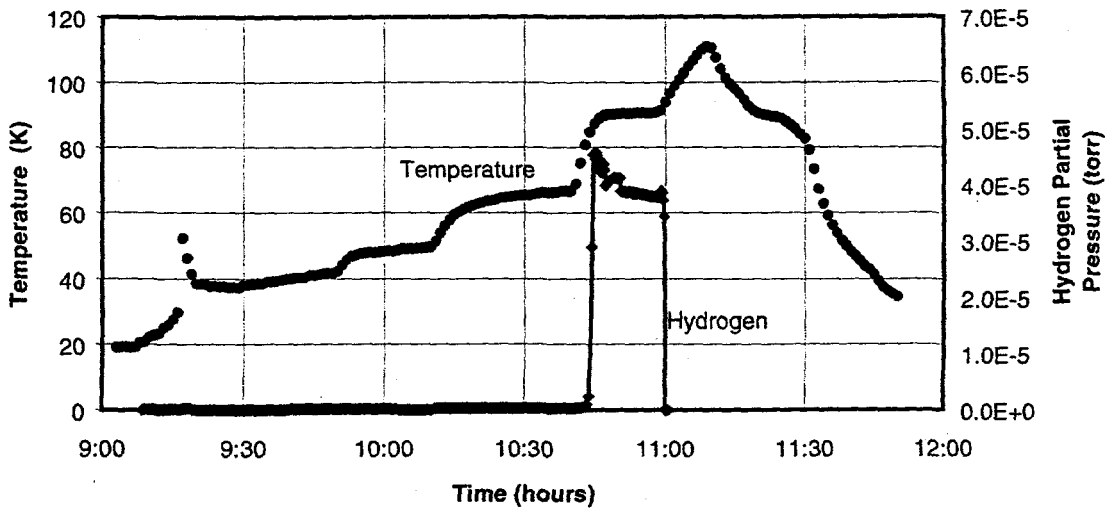

Figure 2. Hydrogen release during moderator anneal.

3.2 "Burping" of Irradiated, Cold Solid Methane

The cause of operational problems in cold solid methane moderators is a radiation induc thermo-chemical instability. Fast neutrons and recoiling protons knock apart methane molecules.

$$CH_4 \rightarrow CH_3 + H$$

Recoiling protons stop in the methane and capture protons from nearby molecules leav further methyl groups and a hydrogen molecule.

$$H + CH_4 \rightarrow CH_3 + H_2$$

The net neutron-induced reaction thus produces at least two methyl radicals and hydrogen molecule. H_2 accumulates in the bulk material. CH_3 's diffuse by thermally-activa proton hopping until they come close to one another, then combine in pairs, giving off heat.

$$CH_3 + CH_3 \rightarrow C_2H_6 + heat$$

A coolant removes the heat produced in the system at a rate proportional to the differenc its temperature from the temperature of the coolant.

The equations below (written in an infinite-medium approximation) describe the dependence of the methyl density n and the temperature T reasonably well.

$$\frac{dn}{dt} = R(t) - K\exp\left(-\frac{\Delta E}{k_B T}\right)n^2$$

$$\rho C\frac{dT}{dt} = P(t) + EK\exp\left(-\frac{\Delta E}{k_B T}\right)n^2 - H\left(T - T_{coolant}(t)\right)$$

In the equations,

$R(t)$ is the rate of production of CH_3, K is the recombination rate constant

$\exp\left(-\frac{\Delta E}{k_B T}\right)$ expresses the temperature dependence of the CH_3 diffusion rate

ΔE is the activation energy for CH_3 diffusion, k_B is Boltzmann's constant

ρC is the volume specific heat (depends on T)

$P(t)$ is the nuclear heating power density

E is the energy given off per CH_3 recombined

H is the heat removal constant (depends on T)

$T_{coolant}(t)$ is the temperature of the coolant.

The equations are very difficult to solve, but numerical solutions exhibit both steady behavior and spontaneous oscillations, depending on the values of externally controlled variables, the coolant temperature, the rate of production of methyl radicals, the heat removal constant, and the nuclear heating power density (which vary with operating conditions), and the values of the intrinsic parameters of the system, ρC, ΔE, K, and E.

3.3 Solid Methane vs. Liquid Methane

During the summer of 1994, IPNS installed a completely new moderator/reflector assembly, a second liquid helium refrigeration system and a new gas control system to convert operation of the H moderator from liquid to solid methane. The installation, completed in September, 1994, included replacing the liquid methane moderator can in the H position with a new solid methane moderator can and installing a new solid methane moderator can in the C position. The new solid methane H moderator contained an aluminum foam heat transfer medium (the same as always used in the C moderator) and an aluminum - 17 wt % gadolinium poisoning plate, 0.5 mm thick by 10 cm square, located 2.5 cm beneath the viewed surface.

Compared to the previous liquid methane H moderator (~100 K), the colder solid methane H moderator (~30 K) produced a greater flux of long wavelength neutrons, although there is some sacrifice of flux in the ~10 meV region, and also extended the sharp slowing down pulse shape to lower energies. This enabled the high resolution (medium energy) chopper spectrometer (HRMECS) which is served by this moderator to extend its useful range to an incident energy (E_0) below 4 meV, where about 150 μeV elastic resolution is achieved. The counting rate in the quasi-elastic neutron scattering spectrometer (QENS) in the near-elastic range (E_t = 3.7 meV) increased substantially with little resolution change. Also, the glass, liquid, and amorphous materials diffractometer (GLAD) enjoys cleaner resolution in the longer wavelength range.

4. CONCLUSIONS

IPNS continues to successfully operate with solid methane as the medium in the C and H position moderators by taking precautions to periodically remove: (1) stored energy that accumulates during operation and is directly proportional to accumulated dose and inversely proportional to operating temperature, and (2) hydrogen (formed by neutron radiolysis) that is trapped in the solid methane. The stored energy and accumulated hydrogen can be released by warming the moderators to approximately 90 K. The annealing causes some methane gas loss and makes a complete change of gas necessary after about one week of operation to maintain neutron intensity.

The understanding of stored energy release "burping" so far is based on relatively poorly controlled measurements on production versions of solid methane moderators. However, it is too

difficult to vary conditions systematically, the installed instruments are too few, and the risk to continuing operations is too great to enable a program of measurements on IPNS that would provide clearer insight into the basic phenomenon. IPNS looks forward to carrying out a program of measurements that will provide a better understanding of the burping phenomenon, provide the basis for improving the design of production moderators and for determining operating temperatures and annealing schedules, and give information on the stresses encountered during operation and annealing.

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