ICANS-XI International Collaboration on Advanced Neutron Sources
KEK, Tsukuba, October 22-26, 1990

RECENT EXPERIENCE WITH IPNS MODERATORS

J. M. Carpenter and T. L. Scott
Argonne National Laboratory
Argonne, Illinois 60439

ABSTRACT

We describe experience with the moderators of IPNS in the two years since the installation of the Enriched Uranium Booster Target.

I. Introduction

The Enriched Uranium Booster Target was installed in IPNS in October, 1988. According to expectations, the neutron beam intensities increased by a factor of 2.5, taken as an average over the three moderators. Accompanying the higher neutron beam intensities are higher nuclear heating densities and correspondingly higher radiation damage rates in the moderators. (The delayed neutron background also increased, but this effect does not directly relate to moderator performance, and is dealt with in another paper in these proceedings.) Anticipating problems on these accounts, which would be especially severe in the cold solid methane "C" moderator, we replaced the methane there with liquid hydrogen; this required no modifications to the system. Therefore since Booster Target installation our moderators have been as shown in Table 1.

Table I.

IPNS Moderators Operating Since October, 1988

Moderator	Beams	Description
"C"	C1-3	Liquid H_2 @ 20K, stationary, grooved, decoupled
"F"	F1-6	Liquid CH_4 @ 110K, circulating, flat, poisoned, decoupled
"H"	H1-3	Liquid CH_4 @ 110K, circulating, flat, poisoned, decoupled

Below, we describe our experiences of various kinds, the means we have applied to understand new behavior we observed, and the measures we undertook to cure problems that arose.

II. Temperature Stability of the Circulating Methane Moderators

The temperature of moderators varies as accelerator beam intensity varies during operation. Transients follow beam startup of course, but occur with some frequency due to accelerator trips even though there is rapid recovery. The reason is entirely trivial, in that the temperature variation ΔT is proportional to the accelerator induced power variation ΔP,

$$\Delta T = \Delta P / (\dot{M} C_p) \tag{1}$$

where $\dot{M}$ is the mass flow rate of methane and C_p is its specific heat. Before Booster Target installation, temperatures fluctuated about ± 5K, which was tolerable for most of the experiments. After Booster installation, temperatures fluctuated more than ± 10K, which was not tolerable. We increased the flow rate of methane $\dot{M}$ and installed heaters in the inlet CH_4 stream controlled from the accelerator current. The increased flow rate stabilized the fluctuations again to about ± 5K. The heaters, which are intended to compensate for accelerator-induced power variations ΔP, have not yet been tested. Further to accommodate the temperature fluctuations and the accompanying changes in the neutron spectrum, we have incorporated monitor spectrum normalization into the data analysis routines of the white beam instruments.

III. Intensity Loss in the "F" Moderator

Over the course of each two-or three-week run, we observed that the overall intensity of the beams from the "F" moderator (<u>not the "H" moderator</u>) decreased by up to 50% or more. Simultaneously, the pressure in the ballast tanks of both the "F" and the "H" systems gradually increased. The wavelength spectrum gradually hardened during these periods. Routine gas chromatographic analyses of samples of gas from the systems showed the well-known buildup of H_2 and of heavier volatile hydrocarbons. These appear as a result of to radiolysis due to fast neutron and gamma ray damage to the methane, for example reaction (a)

$$2CH_4 \xrightarrow{n_f\,,\,\gamma} C_2H_6 + H_2 \ . \tag{a}$$

Table 2. shows the results of gas chromatography on a sample of gas from the circulating loop of the "F" moderator system after one week of operation, starting with a fresh charge of CH_4, with an accelerator current averaging 15. μA, and with the Booster Target.

Table 2.

Molar Concentrations in Gas
from the Circulating Side of the "F" Moderator,
After One Week of Operation at 15 μA.

Material	Concentration
H_2	22.6 %
CH_4	75.2 %
C_2H_4	≤ 0.16 %
C_2H_6	1.5 %
C_3H_8	0.39 %
C_4H_{10}	≤ 0.07 %

The buildup of radiolysis products is substantially faster with the booster target than with the depleted uranium target, as anticipated. The loss of intensity can be traced to the accumulation of H_2 vapor in the system; since H_2 is not highly soluble in liquid CH_4 and furthermore is a vapor at the pressure and temperature of the circulating methane, even a small amount of H2 can exclude a substantial fraction of the methane from the system. If the hydrogen vapor tends to accumulate in the moderator, then the intensity decrease and the spectrum should harden. The pulse shapes should be altered but whether broadened or narrowed depends on whether the voiding in the moderator is uniform (bubbly) or stratified; we did not observe or attempt to observe the pulse shape change. Figure 1 shows the variation of the monitor detector counting rate, integrated in the range $0.5 \le \lambda \le 5.0$ Angstroms, for the F2 (GPPD) beam. Figure 2 shows wavelength spectra measured with the GPPD monitor detector at different times during a running cycle. Although these results are typical, the behavior was somewhat different in different running periods. We argue on admittedly flimsy grounds that the "F" moderator container (inlet at the bottom, outlet at the top) differs from the "H" moderator container (both inlet and outlet at the top) in such a way as to explain that "H" acts like a physical phase separator collecting liquid, while "F" promotes a "bubbly" situation.

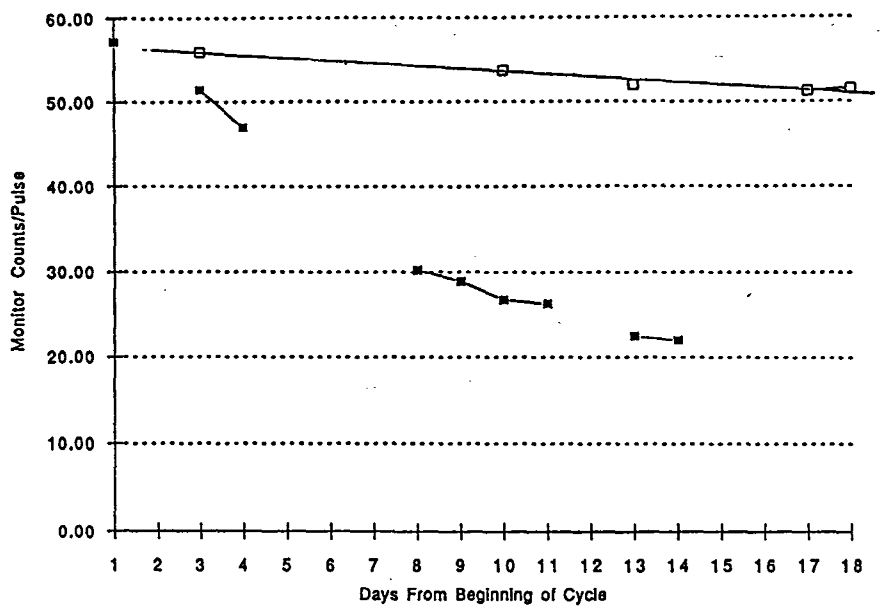

Figure 1. Variation of the intensity of the "F" moderator beam as a function of time into a running cycle. Closed symbols, March, 1990 cycle (no H_2) separator; open symbols, May, 1990 cycle (H_2 separator operating).

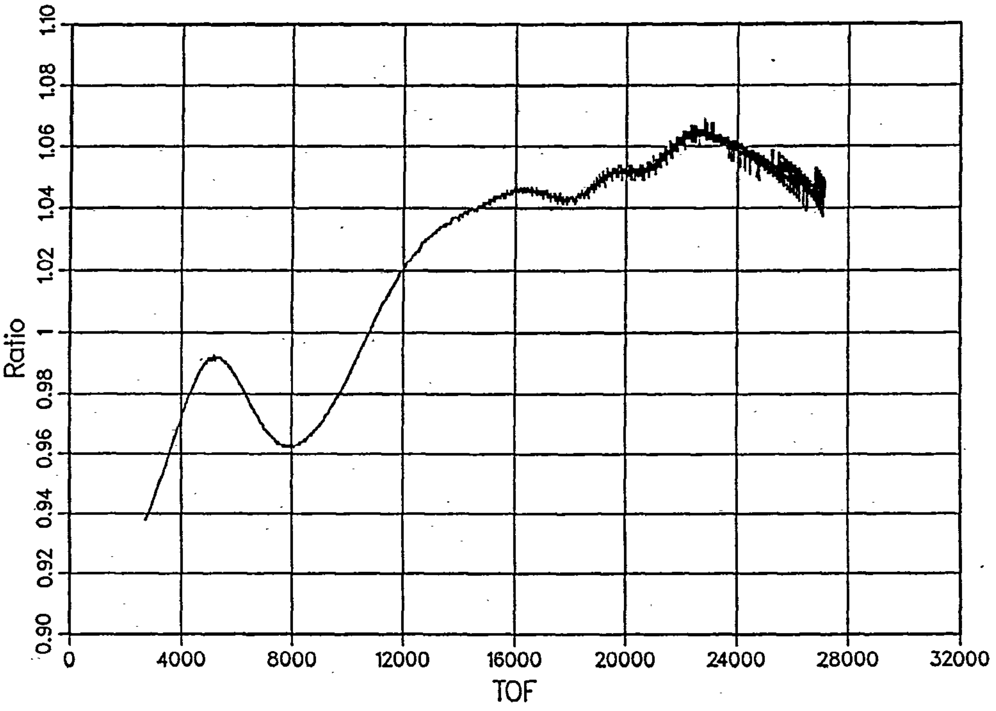

Figure 2. Variation of the spectrum during a running cycle. The figure shows the ratio of monitor counting rates, normalized according to accelerator proton current, at the beginning and at the end of a running cycle. The ratio shows the gradual increase in the temperature of the Maxwellian.

Figure 3 shows a highly schematic diagram of the circulating methane moderator systems.

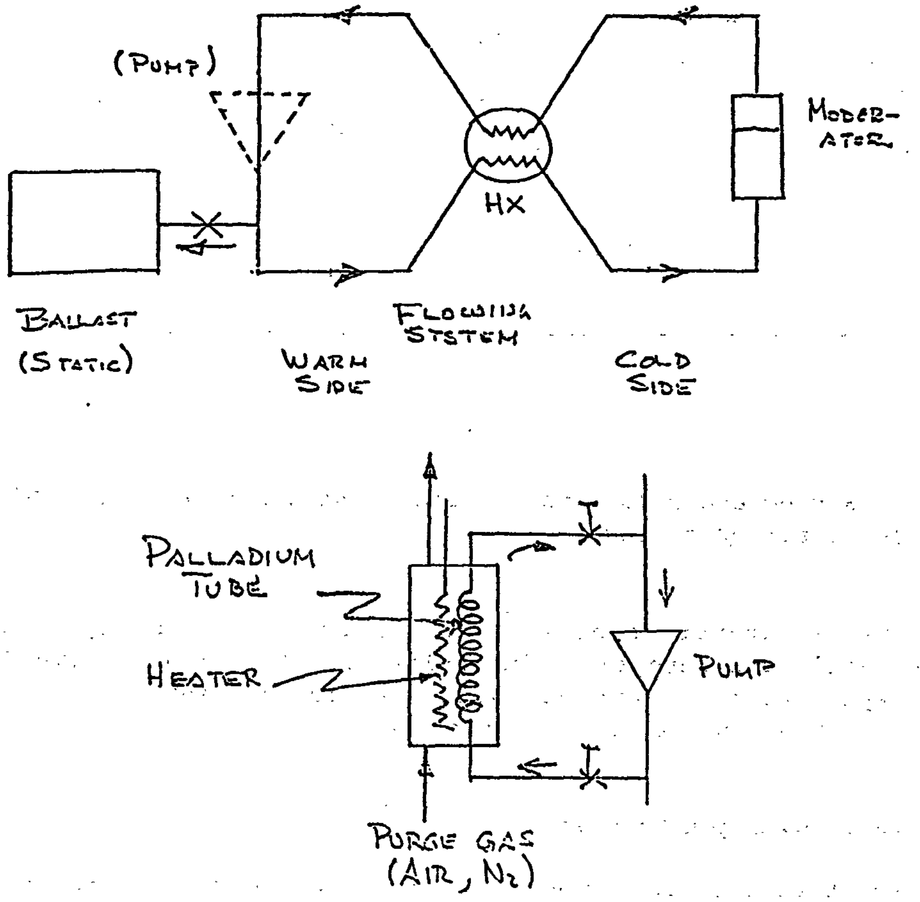

Figure 3. Schematic diagram of the circulating liquid methane moderator systems, and of the H_2 separator.

We developed a numerical model of the system, including the effects of solubility of H_2 in liquid CH_4. Calculations from this model show qualitative agreement with observations, but we have been unable to refine the unknown parameters to provide a truly acceptable match of calculation to measurement. Modeling calculations show that the ballast tank pressure is a sensitive indicator of the behavior of the system.

On the basis of the gas chromatography and numerical simulation results, we installed a commercial hydrogen separator in a bypass loop around the (room temperature) methane circulating pump. The separator, model RSD-50 by Resource Systems, Inc., E. Hanover, New Jersey, consists of a thin-walled, heated palladium tube through which the methane passes; hydrogen preferentially diffuses through the tube walls to the outside where it is removed by a nitrogen stream. The numerical model includes the effect of the separator. Figure 4 shows the calculated and measured ballast tank pressures, with calculations for two assumed H_2 production rates and for the value of the H_2 flow rate for the separator that best matches the data.

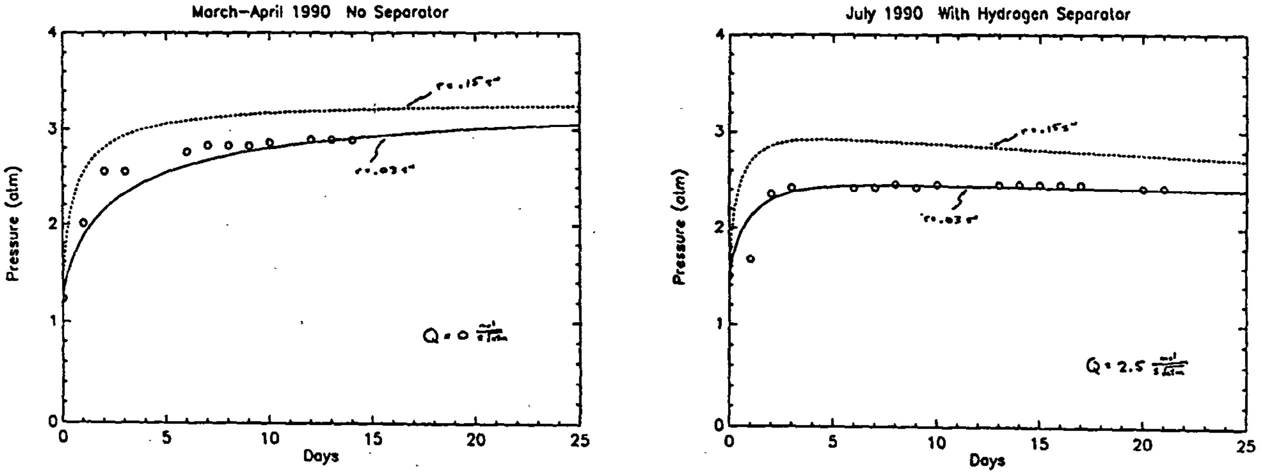

Figure 4. Measured and calculated ballast tank pressures with and without the hydrogen separator operating.

The hydrogen separator stabilizes the operation of the system. Figure 5 shows the wavelength integrated intensity as a function of time during a run with the separator operating; the intensity is constant within about 5% over the course of the two week operating cycle.

IV. The Liquid Hydrogen Moderator

With no essential modifications we now operate the "C" moderator filled with hydrogen. The liquid hydrogen moderator has behaved perfectly. However, the intensity of long-wavelength neutrons has diminished by about the same factor as was gained by installation of the Booster Target, about ×3.5 smaller than would be the case with 20 K methane. For a short time, we operated the "C" moderator filled with 20 K methane, using the Booster Target. Figure 5 shows the ratio of intensities measured with 20 K solid methane and with 20 K liquid hydrogen.

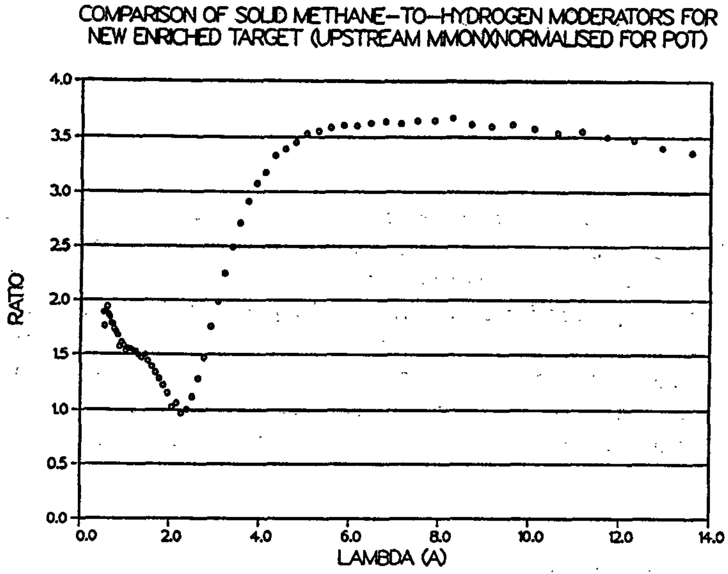

Figure 5. Ratio of intensities of neutrons, normalized to the proton beam current, from the "C" moderator filled with 20 K solid methane to the intensity when filled with 20 K liquid hydrogen, vs wavelength.

The intensity loss is due in part to the lower proton density of liquid hydrogen compared to solid methane, and in part to the inferior low temperature thermalization properties of hydrogen compared to methane. This latter is in turn due to the presence in CH_4 of closely-spaced rotational modes with approximately 1 meV (~ 12 K) spacing; the corresponding lowest energy rotational transition in H_2 is approximately 15 meV (180 K). Although we have made no careful measurements of the pulse widths, the widths of the pulses from the liquid hydrogen is greater than for solid methane at longer wavelengths, as inferred from measurements on the two reflectometers.

Our intent is to return to solid methane filling in this moderator, to recapture the lost intensity and resolution. This will require that we understand the "burping" phenomenon more thoroughly than we do at present, and control it for example by periodic annealing. Since warming the moderator leads to potentially large pressure increases due to the expansion of accumulated, trapped hydrogen, we must devise means to control the pressure increases that follow warming of the moderator, and to remove the accumulated hydrogen.

V. "Tails" on Diffractometer Bragg Peaks

Since the installation of the Booster Target, we have observed roughly 200 µsec-long "tails" on certain Bragg peaks in the powder diffractometers. The tails are about 1 % as high as the main peak and contain up to 10 % of the area, the amplitude varying with the wavelength of the neutrons. In spite of intense efforts based on neutron beam observations, we were unable to conclusively identify the origin of these tails (but see "Post-Conference Notes, below).

VI. Conclusion

The moderators of IPNS have operated more-or-less satisfactorily since installation of the Booster Target. Problems with temperature stability and loss of intensity have been diagnosed and corrected. The 20 K solid methane moderator has been replaced with one of liquid hydrogen, which operates quite satisfactorily, but which provides fewer long wavelength neutrons and poorer pulse width resolution than the solid methane system. Work continues to obtain better understanding of the "burping" behavior of cold solid methane and to control these excursions and the consequent rises of internal pressure.

VII. Post-Conference Notes

A new problem arose immediately after the ICANS Meeting, which was not reported there but we describe briefly here. The "F" moderator methane flow stopped due to a blockage of the system between the outlet of the inlet heat exchanger and the inlet of the outlet heat exchanger, about 10 m of tubing each direction from the moderator itself. Flushing the system with tri n-butyl phosphate - heptane, heptane, methyl ethyl ketone - toluene, and toluene solvents brought out several tens of ml of involatile liquid and some insoluble chips of insoluble solid. We have not been able to identify these materials. The system was reassembled after a week's shutdown for this work, and the moderator operated successfully for one week, after which the system clogged up irreparably. We have since removed the "F" moderator and replaced it with a new, clean assembly, with inlet and outlet arrangements within the moderator container arranged to provide more effective liquid phase separation. Meanwhile we have flushed the heat exchangers and external piping with toluene. The removed materials are being subjected to chemical analysis. Results so far indicate that the solid material is insoluble in any solvent compatible with the construction materials, and is a hydrocarbon with H/C ratio of about 8:5.

The origin of the "tails" on the "F" moderator pulses has been identified; Cadmium void liner above the beam voids on both sides of the "F" moderator were accidentally omitted during assembly of the system, prior to Booster Target installation. These have been provided in the rebuilt assembly.

VII. Acknowledgements

Efforts on the IPNS moderators are pursued by the Cryogenic Moderator Problems Working Group, D. E. Bohringer, J. M. Carpenter, R. K. Crawford, W. C. Dimm, R. Kleb, A. W. Schulke, T. L. Scott and K. W. Shepard.

This work was performed under the auspices of the U. S. Department of Energy, Division of Materials Sciences, Office of Basic Energy Sciences, under contract W-31-109-ENG-38.

Q(G.S.Bauer): With the scheme of a pumped slurry of methane spheres being technically rather complicated, do you think that a pebble bed of methane spheres cooled by liquid hydrogen or even helium would limit the burping to individual pebble and thus spread it out in time without risk of the container bursting?

A(J.M.Carpenter): Yes! The consequent improvement in the effectiveness of cooling also will tend to stabilize the system.

Q(Y.Takeda): In the temperature history, are there any periodicity found, which corresponds to a limit cycle?

A(J.M.Carpenter): Yes, we observed a period of about 1 day under the conditions of IPNS depleted target operation.

Q(Y.Takeda): What are the control parameters of the nonlinear governing equation set for burping?

A(J.M.Carpenter): Principally, the coolant temperature, given that power and damage density are fixed. Otherwise, the heat transfer coefficient between CH_4 and coolant is a useful design parameter, and can be controlled somewhat by regulating coolant flow rate.

Q(R.Pynn): Have you looked at you burp equations for a range of parameters? It would surprise me if limit cycles were the solution for more than a small range of parameters.

A(J.M.Carpenter): In fact we had trouble finding parameters that produce oscillations, before the linear stability analysis. Now we have a guiding theory, which works.