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The IPNS Grooved, Solid Methane Moderator*

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Introduction

There are two motives for using cold moderators in pulsed neutron sources, to provide higher fluxes of long-wavelength neutrons, and to extend the epithermal range with its short pulse structure to lower energies. For both these purposes solid methane, operated at the lowest possible temperatures, is the best material we know of. Two problems accompany the use of solid methane in high power sources, namely heat transport in view of the low thermal conductivity of solid methane, and deterioration due to radiation damage. We have designed a system suitable to operate in IPNS, subject to nuclear heating of about 25 W, which incorporates an aluminum foam matrix to conduct the heat from within the moderator. We report the results of the first few months' operation, and of a few tests that we have performed.

Design of the Grooved Cold Moderator

The goal of the design of the grooved solid methane moderator was to provide as low a spectral temperature as possible, with no constraint on resolution. While the goals need not to have been qualified for the Small Angle Diffractometer, the Polarized Beam experiment which also views this moderator, requires reasonable intensity for 4 Å wavelengths and nearby, and resolution of about 100 μsec. The tests of Inoue, et al.[1] indicate that the spectrum from 10 K methane would not be so cold as to diminish significantly the flux of 4 Å neutrons, and that by a grooved design similar to that now in use at KENS would satisfy the resolution requirement.

However, experience at KENS[2] and our own calculations indicated that with the power densities expected at IPNS, temperature gradients would be too great in pure solid methane. Thus we were led to provide a metal matrix within the solid methane volume to conduct the heat to the cooled walls of the container. Aluminum metal foam seemed to provide the required small subdivision of the methane and the required increase in bulk thermal conductivity, and to be fabricable using well-developed techniques, while the required sacrifice in methane density is small and acceptable. The foam is brazed to the cooled flat surface of the container.

A closed-cycle refrigerator provides approximately 7 K circulating helium gas for cooling. Figures 1 and 2 show the design of the grooved moderator container. A room temperature aluminum can provides the insulating vacuum enclosure. The

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thermocouple is a 0.063 inch (1.6 mm) diameter stainless steel sheathed type T junction. In operation, the moderator is decoupled from the surrounding graphite reflector by 0.05 cm cadmium. The normal mode of operation is to fill the system by condensing the methane into the system at the beginning of a run cycle. At the end of each cycle (typically about 400 hours), after waiting for decay of short-lived isotopes (^{11}C, etc.) we allow the system to warm up and release the gas to the facility exhaust system. Normally we fill the system so that the filling tubing contains an overpressure greater than one atmosphere absolute, with the motive to prevent leakage of air into the system.

Thermal Analysis

Argonne retained the Materials Division of ERG under contract to perform the thermal analysis, design, fabrication, and tests of the Inner Cannister of the Solid Methane Moderator for the IPNS program. ERG was selected because of its unique Duocel Foam Metal which is used extensively in solid thermal mass heat exchangers for various applications such as Infra-red Surveillance Satellites. The material is ideally suited to provide nearly isothermal conditions in the solid methane cryostat.

Doucel foam metal is an open-cell, reticulated, skeletal structure that is manufactured with average cell sizes between 10 and 40 pores per inch (PPI), and density varying from 3% to 12% that of the bulk metal. The pore structure is more or less regular and the ligaments display a purity equal to that of drawn wire or cast aluminum. The conductivity is isotropic because of the uniformity of the matrix.

ERG determined the alloy, density, and pore size to meet the goals of the program. 6101-T6 aluminum foam would provide the best conduction, and would enable brazing to the cooled back plate. The alloy also has superior strength in the heat treated condition. 6101 alloy contains 0.5% Si, 0.5% Mn and other trace constituents, yet retains high thermal conductivity at low temperature. ERG's prior experience with this alloy cycling to low temperature in other applications also indicated its use.

The predominant factor in foam metal heat transfer is the cross sectional area of the ligament. For like densities the coarsest material gives the largest ligament diameter, which means the largest conduction area. For the specified conditions, the mean diameter of the solid methane cells in the 10 PPI material provides for adequate heat transfer from the solid methane to the aluminum ligaments. Thus ERG recommended the 10 PPI foam metal.

The remaining variable in the design is the density of the material required to provide an adequate conductance path for transporting the 25 W of nuclear heating to the back plate. The cross sectional area of the foam ligament varies as the cube root of the density; for example, foam of 3% density has a ligament diameter of 0.012 inches (0.0047 cm), while 6% dense material has a ligament diameter of 0.015 inches (0.0059 cm).

We adopted a worst-case approach to arrive at the required foam density. We found that there is a cross sectional area within the moderator 1.375 in (3.49 cm) high by 1.19 inch (3.02 cm) wide and extending 1.375 inches (3.49 cm) from the cooled back plate, through which all the heat generated within it must be conducted to the back plate. The highest temperature within the moderator would then be expected at this section, 3.49 cm into the foam. Assuming uniform nuclear heating power density we found that this volume would have a total power of 1.067W. With these assumptions we employed a simple one-dimensional finite element analysis to determine the temperature rise.

$$Q = [C_{\mathrm{alum}} \times \mathrm{Area} \times (\mathrm{foam\ density}/2.8)$$
$$+ C_{\mathrm{methane}} \times (1. - \mathrm{foam\ density})] \times \Delta T/\Delta x$$

where

Q = total power in the section,

C = thermal conductivity,

A = area of section,

1./2.8 is the fraction of metal in the direction of the heat flow (only 33% of the ligaments' mass lies in the flow direction). This factor adjusts for the effect of percolation of heat through the random matrix.

ΔT = temperature increment,

Δx = length increment in the finite element analysis.

We used the following thermal conductivities:

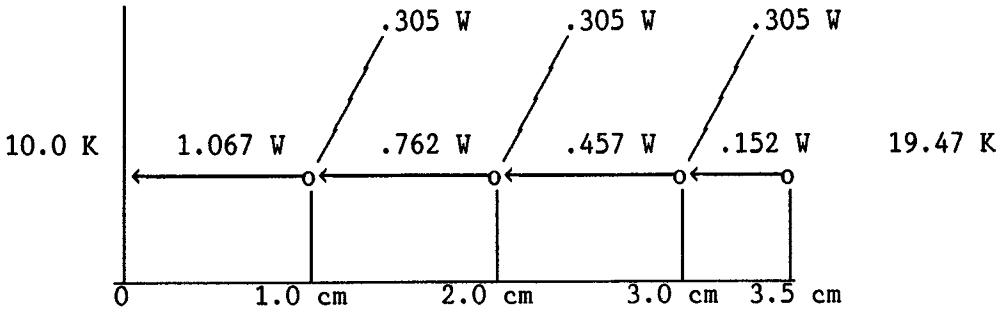
$C_{methane}$ between 10 K and 20 K = 0.003 W/cm-K

C_{alum} for 6101-T6 alloy 10 K = 0.750
11 K = 0.845
13 K = 1.035
15 K = 1.225
17 k = 1.415
19 K = 1.605
20 K = 1.700

For our adopted worst case we found the following temperature increments

Distance	Temperature rise
0.0 - 1.0 cm	5.36 K
1.0 - 2.0 cm	2.48 K
2.0 - 3.0 cm	1.24 K
3.0 - 3.5 cm	0.39 K

which we illustrate below.



The calculation indicates that with a 6 % density, 10 PPI, 6101-T6 Duocel foam metal a maximum temperature of 19.47 K can be expected at the hottest point in the moderator. The moderator actually operates with the full heat load and with helium gas at 7 K, at a temperature of 12 K at the location of the thermocouple.

The Spectrum

We determined the spectrum of the neutron beam emerging from the new moderator, using the "1/v" monitor detector of the Small Angle Diffractometer, approximately 6.4 m from the moderator. Figure 3 shows the resulting counting rate distribution. With the IPNS delivering about 12 µA of 450 MeV protons to the ^{238}U target, the temperature at the location of the thermocouple was approximately 12 K during the measurement. The three lines indicate the locations of aluminum Bragg edges present in the spectrum due to the structural components and the windows in the beam, and verify the wavelength scale. Analysis of the spectrum indicates a Maxwellian temperature 19.9 K, and a thermal to epithermal flux ratio of 4.4 when referred to the slowing-down spectrum in the range 10 - 20 meV. By throttling the flow of cooling helium gas, we briefly operated the moderator with the internal thermocouple indicating 24. K; then the spectral temperature was 36. K and the thermal-to epithermal flux ratio was 5.6. Detailed analysis of the spectrum indicates that it falls below the Maxwellian function due to absorption and scattering of long wavelength neutrons in the beam path, which have not been corrected for in the data.

Operating Experience

The system has operated quite well since its installation in January, 1985. Approximately the expected volume (375. stp liters) of methane gas condenses without difficulty into the container after opening the cooled system to the filled ballast tank, and empties without problems upon turning off the refrigerant flow. However, periodically (about every 30 hours during operation) the system suffers "indigestion", in which the internal thermpocouple temperature rises abruptly to 50 or 60 K, the temperature of the outer can decreases about 10 or 20 degrees from its normal 50 C, and the pressure in the insulating vacuum space (normally 10^{-5} Torr) rises to about 10^{-3} Torr. Mass spectrometer leak detector measurements made during these episodes indicate that the gas in the insulating space is not helium. The radiation monitor on the facility exhaust gas system shows a transient release of ^{11}C, which returns to normal in about 3 minutes.These events (except one, which occurred within a few minutes after shutdown) have occured only during full power operation. We suspect a leak in the methane system, which opens due to the release of stored radiation damage energy. Normally the system recovers from these episodes within about 5 minutes, so that they have no noticable effects on the measurements. After two months' operation, the system developed a leak which appears when the system is warm, but closes when the system is cold. Since that time there have been several major lapses of performance in which the problems persist for several hours and some methane is lost from the system; except for their duration, these appear like the brief events, and have led us to design a new system which will be installed during the summer, 1985 shutdown.

Transient Tests

To determine the response time and the temperature change due to nuclear heating, we recorded temperatures for a period of steady operation before shutting down the acceleartor, and following shutdown. The resulting transient relaxed in an exponential fashion with a time constant of approximately one minute, while the temperature change was approximately 1.K for prior 12 µA, 450 MeV operation. The measurement cannot be considered definitive, since the temperature recording device had a digitizing error only slightly smaller than the change in thermocouple emf.

Radiation Deterioration

Following one 15-day-long cycle, during which the synchrotron delivered 6.5×10^{19} protons to the ^{238}U target, we collected the volatile products in a previously-evacuated tank, following the warming up of the methane. Table I gives the results.

Table I

Analysis of Volatile Products of Radiolysis of
Methane from the IPNS Solid Methane Moderator

Product	Mole fraction* of gas, %
H_2	5.38
CH_4	91.37
H_2O	<0.04
C_2H_4	0.20
C_2H_6	2.30
N_2	not determined-used as mixing gas
O_2	0.17
Ar	0.04
C_3H_8	0.42

* Exclusive of N_2. Average after two periods of mixing, 45 min and 100 min.

The results indicate substantial deterioration of the material, and do not include whatever gases remained in the tubing between the moderator and the collecting tank, nor any non-volatile products. These findings are roughly consistent with those for the KENS solid methane moderator,[2] but our product yields are somewhat smaller, in spite of the fact that the IPNS ^{238}U target produces about twice as many neutrons per proton as the KENS W target. This may have to do with the coupling between the KENS moderator and the KENS target, which is superior to the target-moderator coupling for this moderator in IPNS.

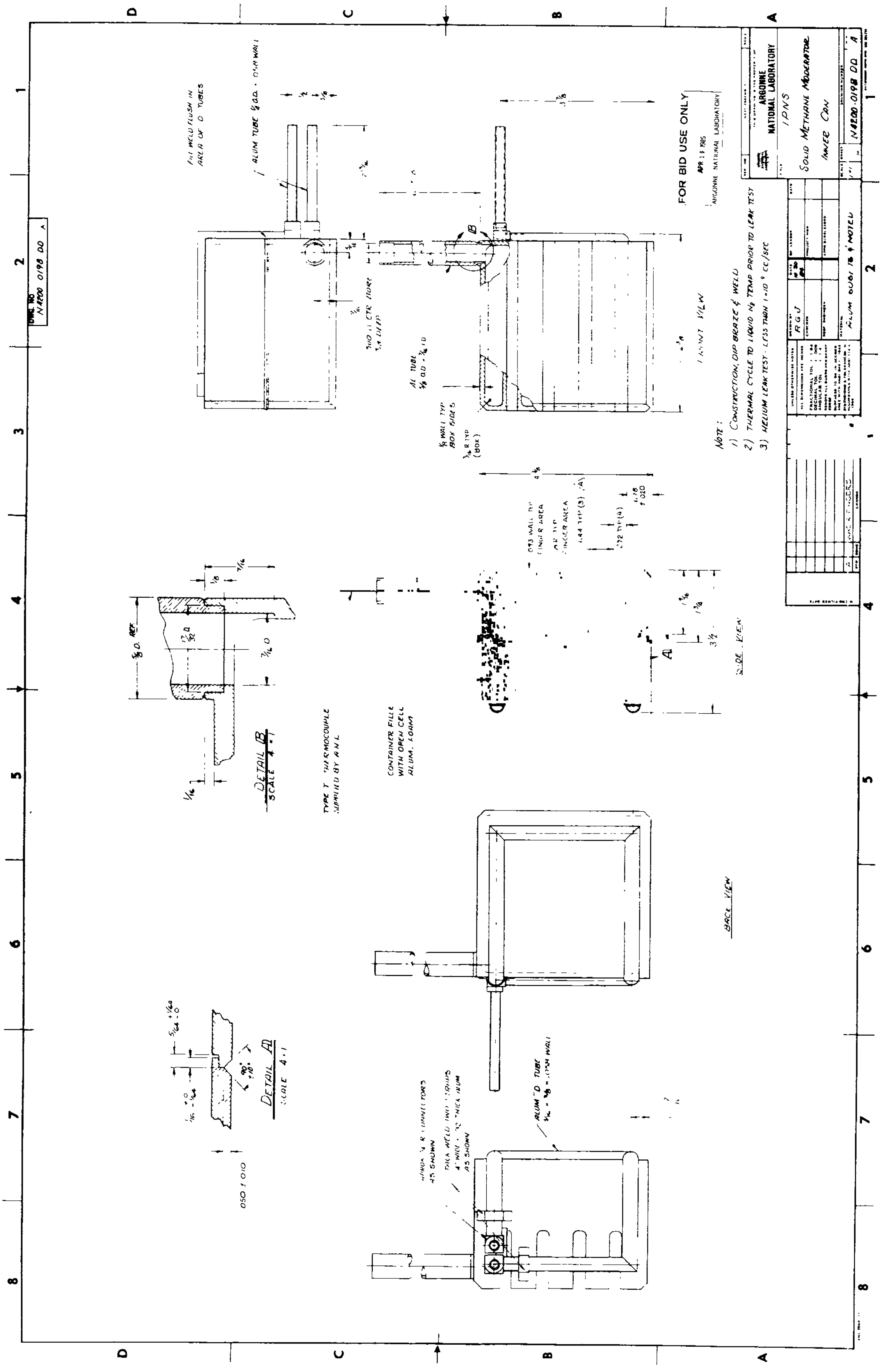
Conclusions

We have developed a successful low temperature solid methane moderator, which operates under conditions of higher power density than can be accomplished in pure solid methane. The measured spectral temperature is the lowest that we know of from all cold neutron sources in existence.

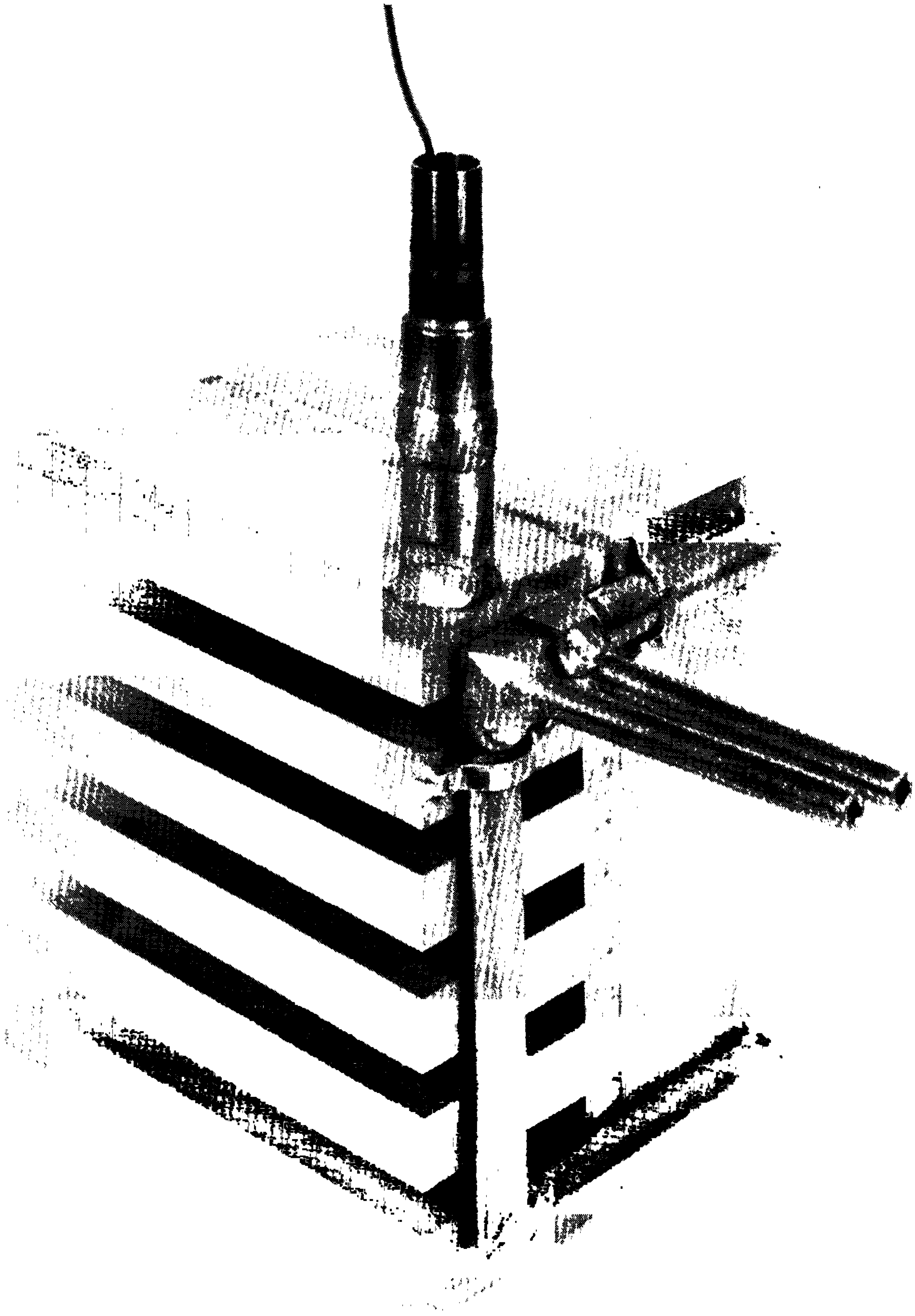
References

[1] K. Inoue, Y. Kiyanagi, H. Iwasa, N. Watanabe, S. Ikeda, J. M. Carpenter and Y. Ishikawa, ICANS VI, p 392, (1983).

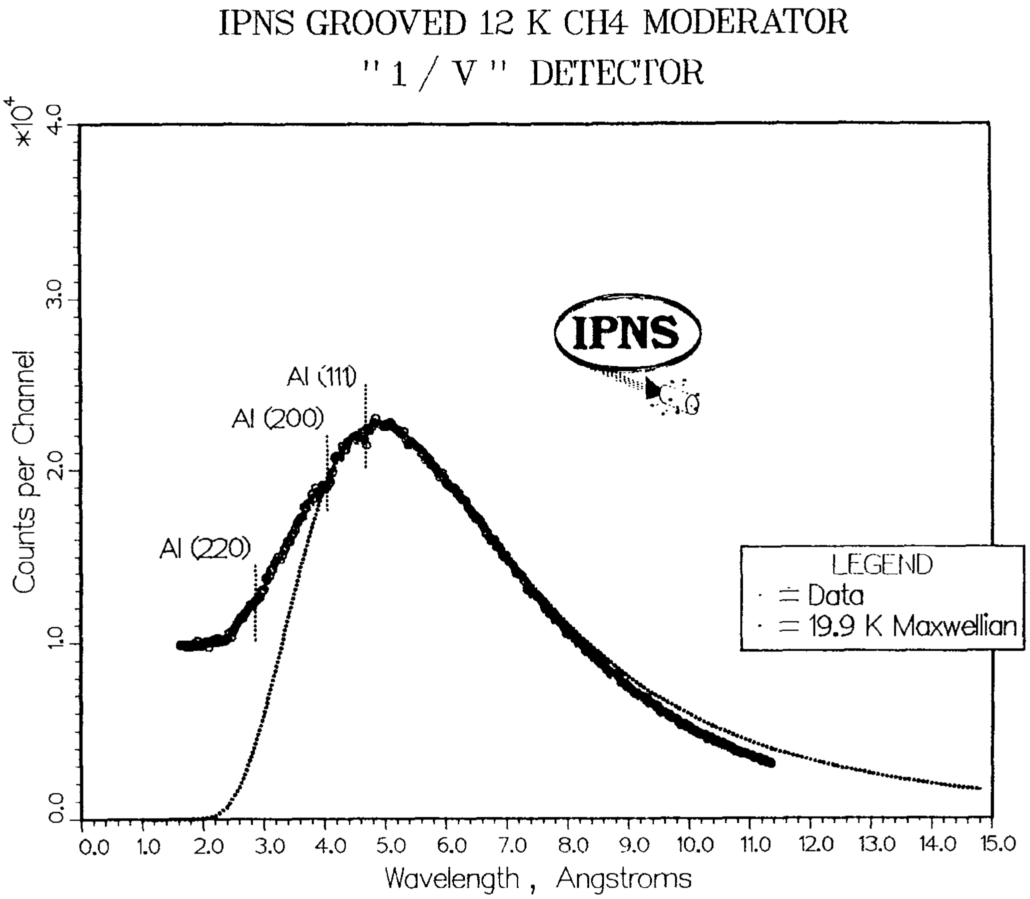
[2] Y. Ishikawa, S. Ikeda, N. Watanabe, K. Kondo, K. Inoue, Y. Kiyanagi, H. Iwasa, and K. Tsuchihashi, ICANS VII, p 230, (1984).



1. Design of the inner cannister of the grooved solid methane moderator.



2. Photograph of the inner cannister of the grooved solid methane moderator.



3. The counting rate distribution measured with a "1/v" detector in the beam from the grooved solid methane moderator. The maximum temperature in the moderator at the time of the measurement was 12 K. The dips in the spectrum are aluminum Bragg edges due to structural and window components in the beam.