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First experience with the new solid methane moderator at the IBR-2 reactor

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Abstract

In the 1999 Fall the solid methane moderator (CM) has been installed and tested at full power at the IBR-2 pulsed reactor. Its main features are a beryllium reflector and a light water premoderator. Radiation load on the methane was three times as much as that of IPNS facility, namely, 0.1 W/g. Effects of temperature, operation time, concentration of a hydrogen scavenger, and annealing procedure on both neutron and service performances were studied. Maximum operation time of a newly loaded portion of methane was 4 days. In this time around 30% of methane is transformed into hydrogen, ethane, and high molecular hydrocarbons, and yet no deterioration in cold neutron intensity was detected.

Among new knowledges, the most important are two facts observed: two-fold decrease in hydrogen formation rate when methane is poisoned with 2.5% to 5% of ethylene, and low formation rate of solid, inremovable products of radiolysis – $(1.5\div3)\ 10^{-7}$ g/J, which means that after 10 years of operation the methane chamber will be filled with only 100 g of residue.

Gain of factor 20 in cold neutron flux was obtained as compared to the routine grooved light water moderator. Presently, it is the highest among the intense pulsed neutron sources.

Introduction

The IBR-2 high flux periodically pulsing reactor offers a means for investigation the field of condensed matter physics. The reactor has been in routine operation since 1984. The source of slow neutrons is room temperature light water moderators which provide a peak density of a neutron flux at a moderator surface of up to 10^{16} n/cm·cm/sec at a pulse frequency of 5 Hz.

To extend the experimental capabilities of the reactor, the decision was once made to design and construct a solid methane cold moderator which would enhance the cold neutron flux (at $\lambda>4$Å) by a factor $20\div30$. According to design calculations, a gain factor of $2\div3$ in the cold flux can be obtained by using solid methane at 20K under the IBR-2 operating conditions as compared to liquid hydrogen. In addition, it is safer and more trouble free in operation.

In spite of the fact that the use of methane present certain difficulties (due to its vigorous radiolysis with the release of hydrogen and the storing of energy in radicals) solid methane moderators were operating rather successfully at Japanese and US neutron centers.

In 1992, the prototype of a solid methane cold moderator (SMCM) was constructed and installed near the reactor core in place of a grooved water moderator for experimental channels 4 to 6 [4-7]. It had operated for about 270 MWh at the reactor power of 2 MW before the damage of the methane chamber forced us to remove the moderator for inspection.

The results of operation and investigation of the SMCM prototype are used as a basis for creation of a regular moderator for the IBR-2 reactor.

In 1995, the project for creation of a regular moderator for the IBR-2 reactor was elaborated and in 1999 the regular cold methane moderator which was constructed in 1995-1998 and installed near the core of the reactor and tested at full power in 1999.

Characteristics of the cryogenic moderator for the IBR-2 reactor:

Moderating material	methane (CH_4)
Cooling substance	helium gas
Moderating material of a premoderator	light water
Thickness of a water chamber	90 mm
Volume of a methane chamber	1.2 litters
Thickness of a methane chamber	25 mm
Calculated working pressure	0.6 MPa
Temperature of methane in the operational "Cold" mode "Warm" mode	 30-35 K 60-70 K
Operation life (for the reactor power up to 2 MW)	5000 hours

The brief description of the cold moderator

The cryogenic moderator (See Fig.1) consists of

a) water premoderator,
b) methane cavity,
c) helium heat exchanger,
d) vacuum jacket,
e) beryllium blocks.

The water premoderator reduces generation of hydrogen in methane considerably, with only moderate loss of cold neutron flux density.

The methane-helium chamber is the determining unit of the whole structure. The vertical and horizontal cross sections of the methane-helium chamber are the squares 240 mm by 240 mm.

The outer side of the methane-helium chamber has six 25 mm high edges of rigidity on which 12 beryllium blocks hang.

The basic purpose of the beryllium blocks is to double the neutron flux due to reflection of fast neutrons. Heat, the result of radiation heating of the beryllium blocks, helps to form a slit gap between solid methane and the aluminum wall of the methane chamber.

This gap of 2-3 mm creates most favourable conditions for release of radiolytic hydrogen from solid methane and prevents its accumulation inside the chamber.

Inside the chamber are 492 heat conductors made from pure aluminum. The heat-conductors are the pivots with the diameter 2 mm and the height 23 mm in methane which

have a threaded connection with the wall of a helium heat-exchanger. The heat conductors are arranged with a step 10 mm.

On the upper side wall, the methane chamber has a pipeline for introducing methane into the chamber and the release of gases.

The walls of the methane-helium chamber are made from aluminum alloy. The thickness of the outer wall relative to the neutron flight direction is 5 mm.

Over the volume of the methane chamber there are 9 thermocouples for the continuous control of temperature in the operational mode in the interval from 10 to 350 K.

The helium heat exchanger maintains the necessary thermal mode of operation of the cryogenic moderator. The heat exchanger is made from aluminum alloy with a comparatively high thermal conductivity at cryogenic temperatures, i.e., 270 W/m·K for 20 K. In the heat exchanger, cooling helium is divided in two equal flows which move upwards along slit flutes in two central sections cooling the wall of the methane chamber as well as methane by means of heat-conductors. Then the flows turn by 180°, move along the slit flutes in the peripheral sections of the heat exchanger and leave the exchanger through outlet pipes.

The simplified flow diagram of the CM is shown on the Fig 2.

Description of the modes of operation

It is planned to use the cryogenic moderator for 3 reactor cycles a year.

The reactor cycle is 250-280 hours. Thus, the operation life of the moderator is 6-7 years. Then, it must be put in a storage.

The modes of operation of the cryogenic moderator at the reactor power 2 MW are:

1) The "warm" operational mode. The temperature of methane is 60-70 K. The mode lasts 120 hours. Then, methane is reloaded.
2) The "cold" operational mode. The temperature of methane is 30-32 K. The mode lasts also 120 hours, but with 4÷5 annealing when the methane temperature is held 65-75 K for an hour. During annealing radiolytic hydrogen diffuses from the methane chamber.
3) The operational mode without methane in chamber; the water premoderator at 300 K serves then as a thermal neutron source. The walls of the chamber are cooled with gaseous helium filling the vacuum jacket of the chamber.

Operational parameters for operation of the cold moderator

The test period spanned a total three reactor cycles. The reactors power in cycles varied from 0.2 to 1.5 MW, and the methane temperatures range was from 30 to 70 K. The total energy obtained by the moderator during the test was 731 MWh.

Cold moderator operated for 216 hours at 30-40 K and for 242 hours at 60-70 K. Also, CM was tested 295 hours without methane in chamber at the 1.5 MW power.

In all seven methane loading were condensed, six of which were of 32 mole, and one of 22 mole. The main tasks to have had to be solved in the tests were:

- measuring of neutron characteristics of the cold moderator,
- determination of the effect of methane doping with ethylene to reduce hydrogen production rate,
- determination of the possibility of CM operation without methane in chamber,
- assessment of the accumulation rate of solid radiolisis products.

Neutron measurements

The neutron measurements were performed using spectrometers: YMO (beam 4), FDVR (beam 5) and DN-2 (beam 6).During the trials in the reactor power mode, the spectral gain factor in the neutron beam intensity was measured compared to the water grooved moderator (Fig.9). Figure 4 depicts the spectra measured for three different CM states: T=30 K, T=60 K, T=300 K (without methane).In addition to shifting of spectrum maximums towards larger wavelengths as the temperature degreases, a specific characteristic is a strong eat- away in the vicinity of beryllium boundary (λ=3.96 Å, λ=3,58 Å, and λ=3,46 Å). Figure 5 illustrates the G-factor at transition from warm to cold state In the wave range from 1 Å to 1.8 Å G<1, from 2 Å to 4 Å grows from 1 to 10, and at λ=6.5 Å it reaches 20. A comparison of spectra from a CM prototype and a regular CM is shown in Fig.6. It is seen that the regulur CM gives a noticeable gain at λ<2.5 Å and also at λ>4 Å; and also at λ>8 Å for which the gain reaches 4.

Radiolytic hydrogen production rate

This value was estimated by measuring of pressure in a receiver builded up with a released hydrogen. For a pure methane, hydrogen production rate was found to be:

$$\frac{dM}{dQ} = (0{,}93 \pm 0{,}03) \cdot 10^{-3} \cdot M(1 + e^{-Q/Q_0}) \qquad (1)$$

where $\frac{dM}{dQ}$ is an amount of molecular hydrogen (mol) produced per 1 MWh of reactor operation; M is mass of methane (mol) loaded; Q is a value proportional to a burn-up of methane, or, to an energy generated by the reactor, MWh; Q_0=(55±5) MWh.

In the 5-th, 6-th and 7-th runs the chamber was loaded with a mixture of 95%-97.5% methane and 2.5%-5% ethylene. The results of doping with ethylene on hydrogen are illustrated in Fig.10. The upper curve shows the release of hydrogen from pure methane. Bottom curves shows the release of hydrogen from methane with additives of ethylene. It is seen that reduced production rate (by~50%) occurs only over a period of ~30 hours after condensation of a fresh methane+ethylene mixture. Effect of doping is insensitive to the concentrations explored.

Assessment of accumulation rates of solid radiolysis products

The amount of solid residue was determined by the method based on the extraction of volatile radiolysis products. According to the literature, the main components are high-molecular saturated hydrocarbons such as C_{20} and those higher up to polymers. The radiation release of volatile products (mainly hydrogen) from such substances is G = 2-4 molecules/100eV. For polyethylene, it is G = 3 or ~$3 \cdot 10^{-7}$ mol/J. During the CM start-up, after methane and low boiling-temperature products evaporated and high boiling-temperature products were pumped out, the chamber was cut off the receiver and the solid residue was subjected to irradiation at reactor power of 1.5 MW and temperature in the chamber of 340-350 for 1 to 2 days. Observing changes of pressure in the chamber and tubes it is possible to assess the amount of solid residue assuming that it is polyethylene with a formula:

$$M(g) = \frac{(V/T)_{eff} \cdot dP/dt}{R \cdot G_{CH_2} \cdot q_V \cdot 3600} = 5 \cdot 10^3 \frac{dP}{dp}, \quad (2)$$

where $(V/T)_{eff}$=0,067 l/K is the effective ratio of chamber + tubes volume (which is 9.1 l) to temperature,
R=0.08055 l·atm/K is the gas constant
G is defined above, q_v is a radiation dose rate in polietylene (0.06W/g)
DP/dt is the growth rate of radiolytic gas pressure in the chamber (atm/hr).

Under the condition that the chamber is pumped out for 10 to 12 hours after methane evaporates in every loading, the accumulation rate of solid resins appeared to be of the order of 2 g per 4 days of irradiation, i.e., 12 g per one year of CM operation. If the chamber is not pumped out the accumulated amount of resins will be about 100 g per year.

The final product, carbon black, will be accumulated in the chamber at the rate ~ 10 g/year (in 3 reactor cycles of CM operation)

Conclusions

A new solid methane cold moderator installed at the IBR-2 reactor has provide substantial gains over the routine water grooved moderator and previous prototype of the cold moderator.

Presently, experiments on study of CM performances is proceeded. More detailed investigation of effects of scavengers are planned with a broader range of concentration. In addition, optimal amount of the methane in chamber will be defined to make the cold neutron flux the highest.

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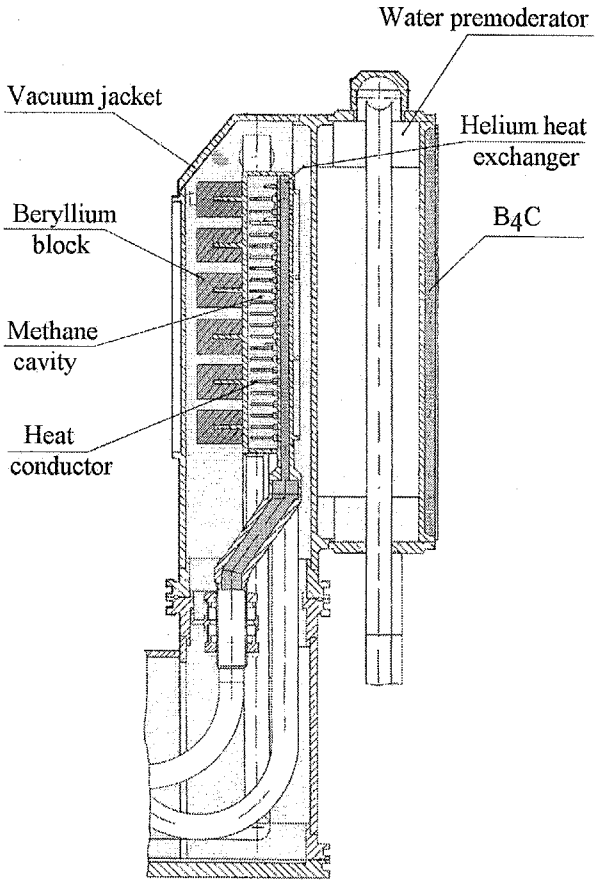

Fig.1. Solid methane moderator section view

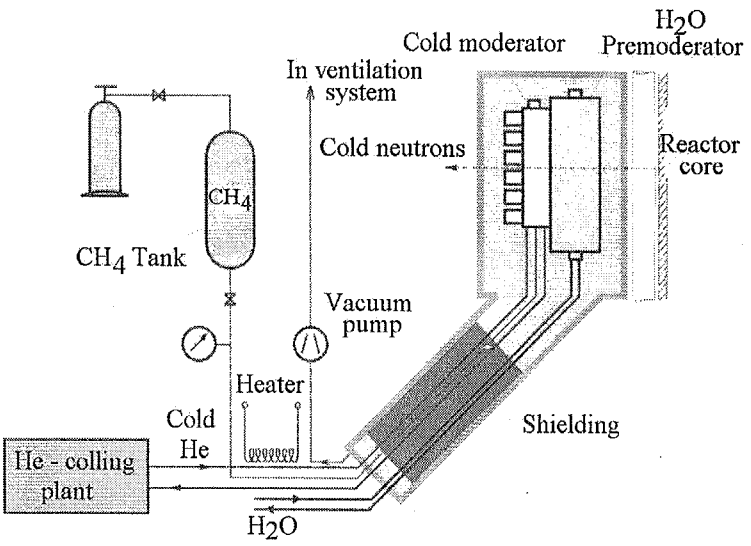

Fig.2. Simplified Flow Diagram of the CM.

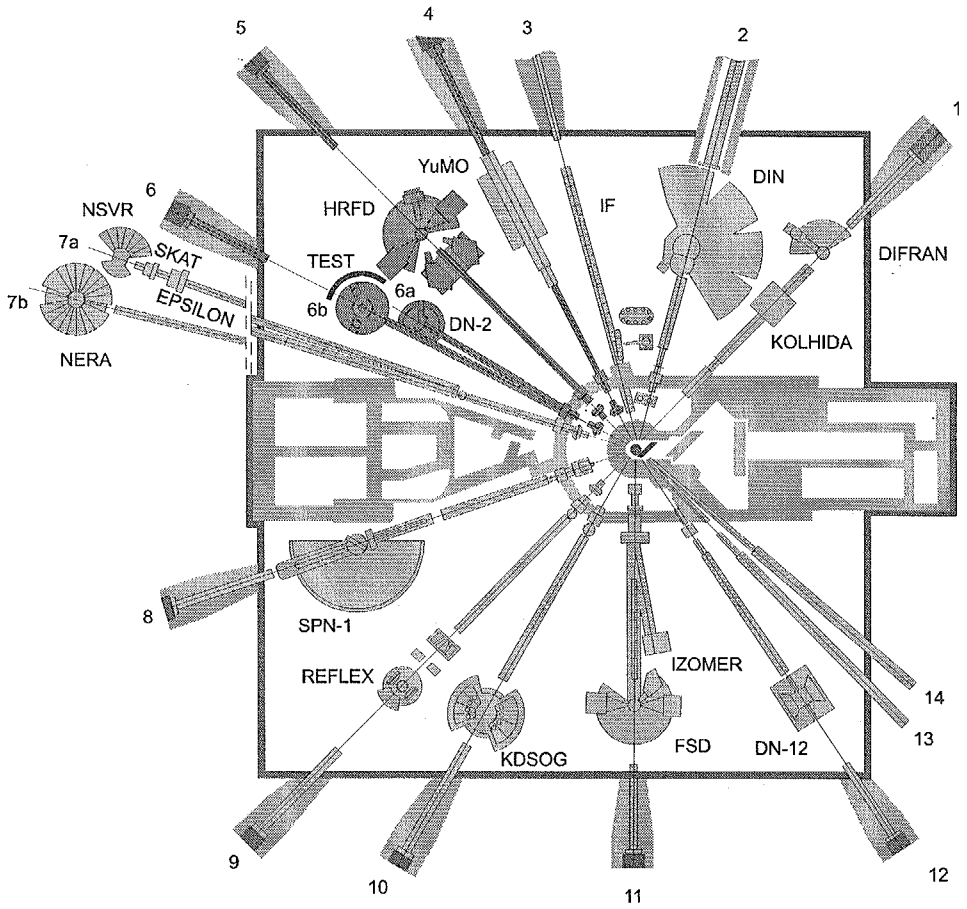

Fig. 3. Layout of the IBR-2 experimental halls.

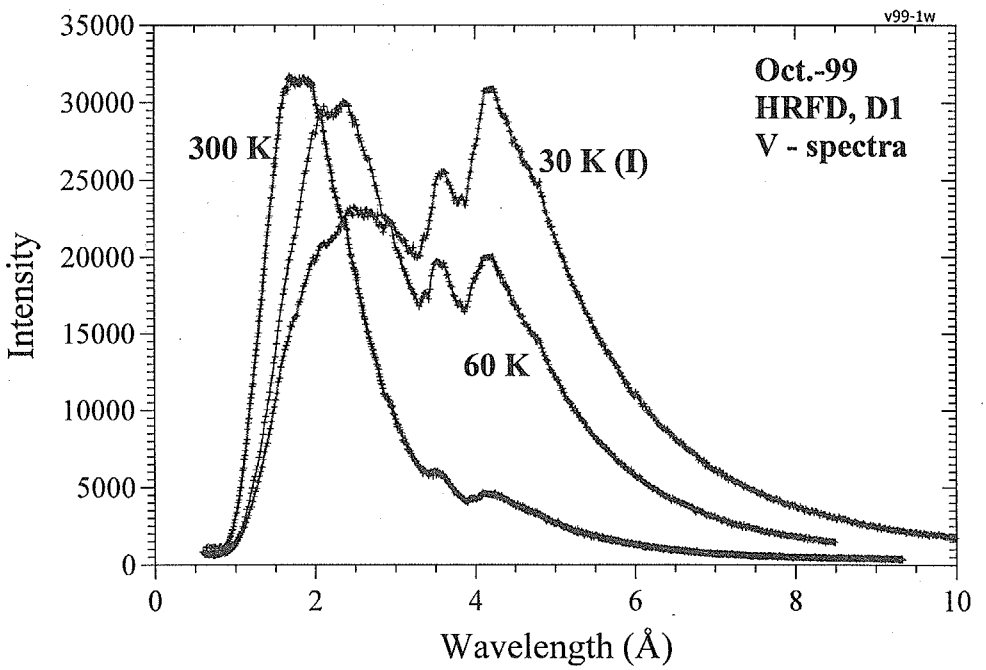

Fig.4. The spectra measured at three different states of the moderator: T = 30 K, T = 60 K, and without methane at temperature of the water premoderator.

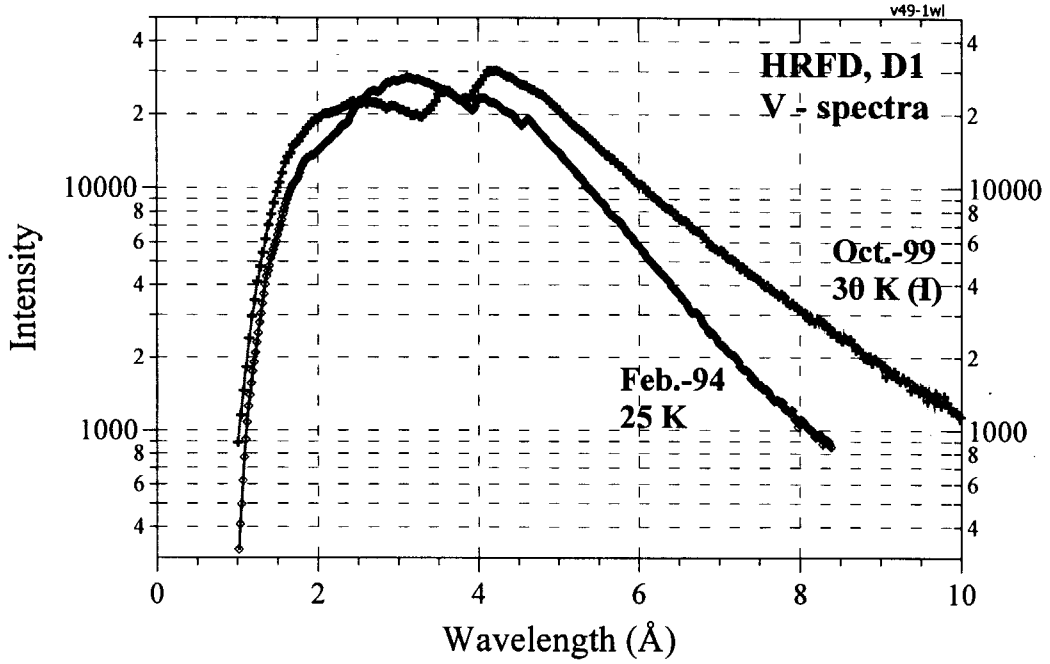

Fig.5. A comparison of spectra of a moderator prototype (year 1994) and a regular cold moderator (year 1999).

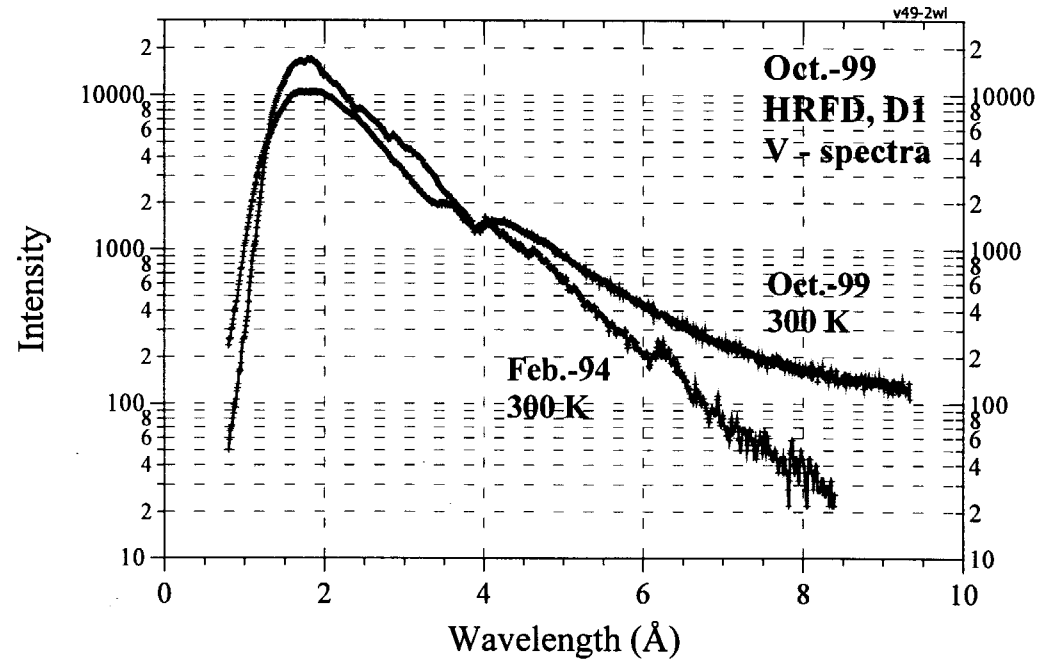

Fig.6. A comparison of spectra of premoderator prototype (year 1994) and a regular cold modulator (year 1999)

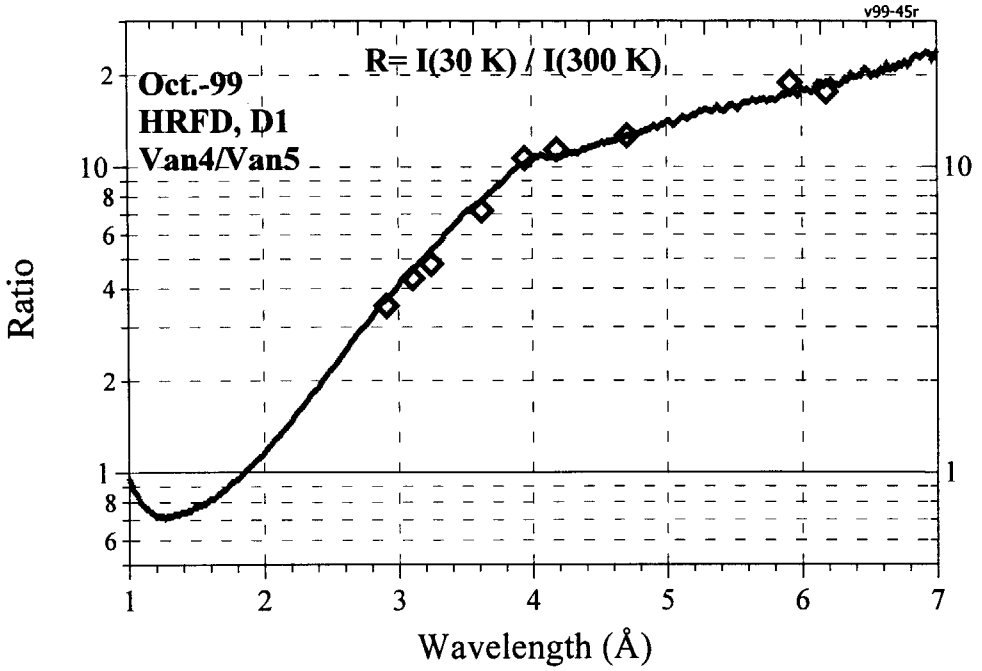

Fig.7. The gain factors of CM in comparison with the premoderator.

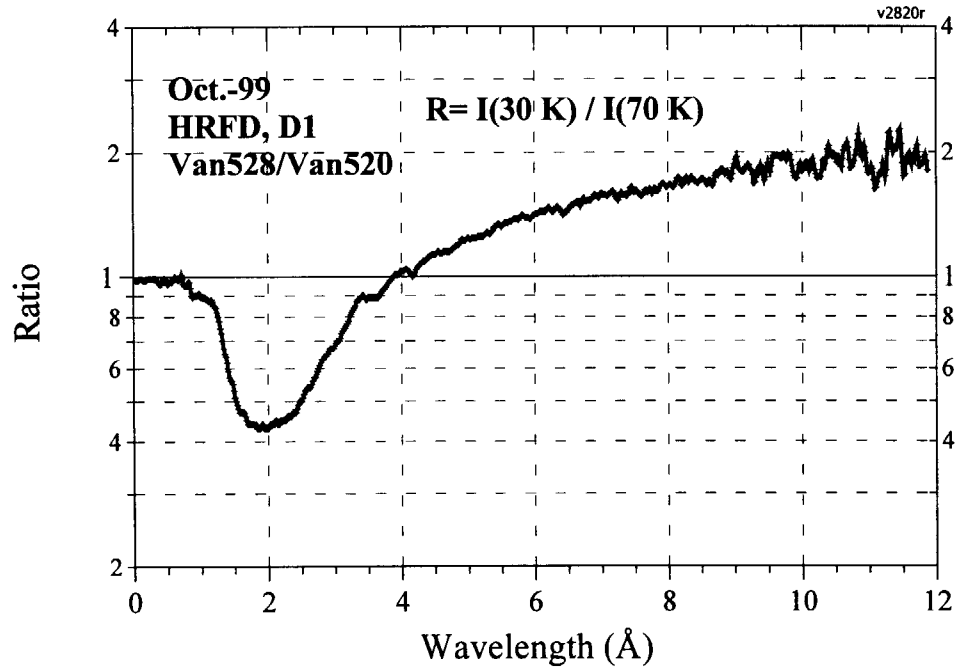

Fig.8. The CM gain (loss) factors at its transition from T = 70 K to T = 30 K.

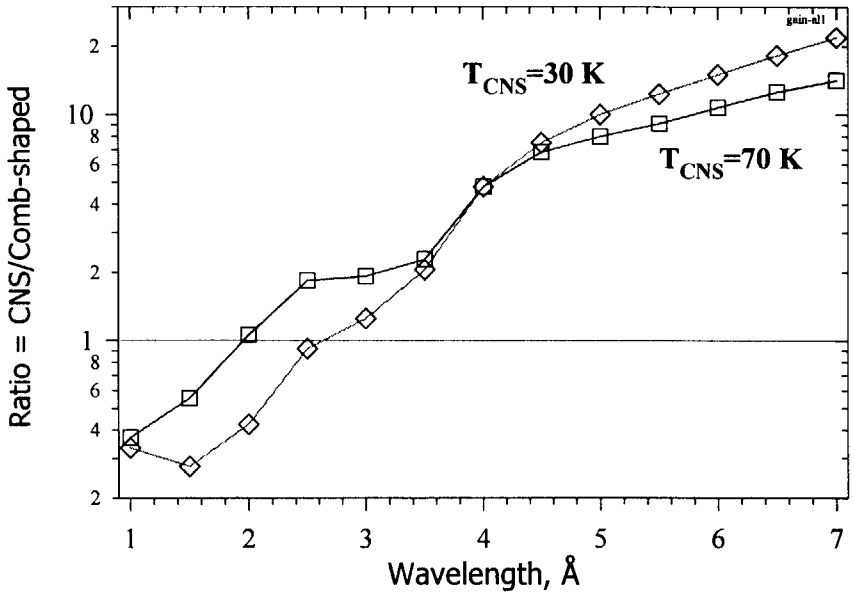

Fig.9. The neutron intensity gain factor for solid methane moderator compared to light water ambient temperature moderator

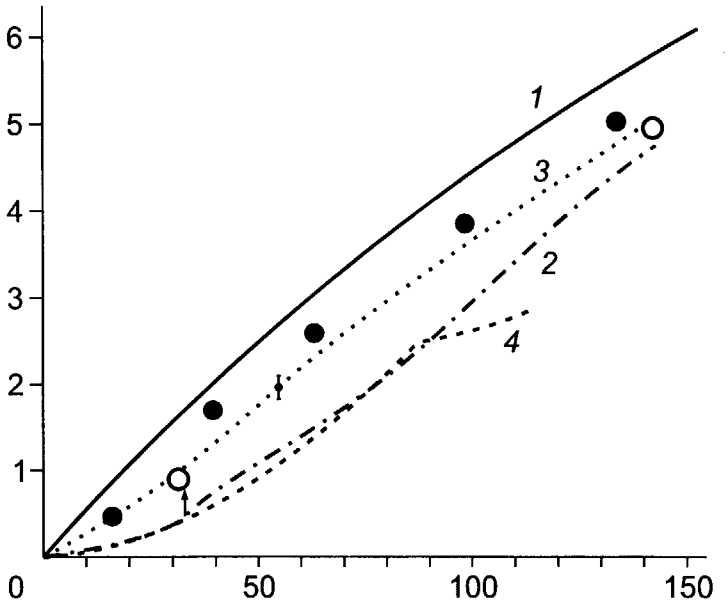

Fig.10. Radiolityc hydrogen release (mol) as a function of energy generation (MWh) from the moment of methane loading (or radiation dose: 1 MWh=0.163 MGy)

1 - the hydrogen accumulation rate in pure methane as obtained from the results of the 1-st, 3-rd, and 4-th loadings;

2 - spontaneous release of hydrogen from methane at ~65K, the 5-th loading with an admixture of 2.5% ethylene;

○ - hydrogen accumulation in the 5-th loading (release during annealing);

3 - the same interpolation and extrapolation of the experimental points;

● - hydrogen released in the 6-th loading during annealing;

4 - spontaneous release of hydrogen from methane at 66-67K, 7-th loading with an admixture of 5% of ethylene (the bent in the curve at 88 MW·h is due to partial evaporation of methane at shut down of He-cooling plant).