FIRST EXPERIENCE OF COLD MODERATOR OPERATION AND SOLID METHANE IRRIATION AT THE IBR-2 PULSED REACTOR

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ABSTRACT

A device comprising three solid methane-containing chambers cooled by 15-60K helium was installed near the reactor core of the IBR-2 reactor. Heat deposition due to irradiation was about 0.12 W/gram. That is a few times higher than earlier experienced at ANL and KENS SNS. A number of effects, such as hydrogen production rate, temperature and pressure dependence of hydrogen release, swelling of methane, thermally induced chemical reactions of radicals recombinations ("burps") were carefully investigated.

The main result was that solid methane can be used as a cold moderator material under the strong neutron irradiation conditions that the IBR-2 reactor displays. It can withstand 4-5 days of continuous operation without recharging, if hydrogen is released periodically. The only limitation is decrease of cold neutron intensity.

Additionally, the cold moderator itself was installed at the IBR-2 reactor to test its neutronic performances. The cold neutron intensity gain, compared to the light water grooved type moderator appeared to be a factor of 20-30. That is in agreement with predictions. The solid methane moderator will begin routine operation as soon as it is commissioned.

INTRODUCTION

It is a fact that the solid methane moderator being installed at a pulsed neutron source offers the highest cold neutron flux /1-3/. For SNS the gain factor is considered to be 5-6 if compared with a liquid hydrogen moderator. For the IBR-2, the gain factor is estimated to be as much as 3. But, developing the CH_4-moderator for IBR-2 was a very difficult task due to the high value of fast neutron irradiation that is on the order of 10^{13}n/cm^{2}sec. We could not have overcome the problems of radiolytic hydrogen generation and thermal explosion of radicals if we had not learned from the experience of American and Japanese scientists with sCH_4-moderators.

The IBR-2 CH_4-moderator (abbreviation is CM) was fabricated in 1991-1992; the construction features and predicted performances have been explained at the last ICANS-XI in Tsukuba /4/. Neutron performance of the CM was measured during a short-term testing run in November 1992. In December 1992 - January 1993 the complex programme of investigation of CH_4 resistance of radiation was pursued ("URAM experiments"). Irradiation conditions were consistent with those for the standard operation of the CM.

This paper deals with both the CH_4 irradiation investigations and the short-term test operation of the CM.

TEST RUN OF THE IBR-2 COLD MODERATOR

The CM was installed behind a 75 mm thick, light water premoderator. Methane was condensed and cooled with a cryogenic machine HGU-500 with 500 W of cooling power, before the reactor was started up. Power of the reactor was raised step by step to 1 MW, 50% of nominal power. At each step, the cold neutron flux was measured by four experimental groups. The CM was operated for 5 MW-hours. The gain factors of the CM to the customary light water, grooved - type moderator are in Fig 1. Measured neutron fluxes match the predicted values (see Table 1).

Temperatures inside methane were 16 - 20 K (at 0.5 and 1 MW, respectively). At 2 MW it was expected to be 27K. Radiation heat production in the methane chamber at 1 MW was 180 W, about 50W was due to neutron moderation. The lag time free thermometer, based on neutron spectrum measurement during one period of reactor pulsation, appeared to be the perfect instrument. Neutron spectrum does not fit the Maxwell shape well, but the closest approximation gives 31k.

After the analysis of reliability of the CM operation based on the URAM-experiments results is completed, the CM will be put into permanent operation, presumably, in the Fall of the current year.

Table 1. NEUTRON FLUX FOR CH_4-MODERATOR, IBR-2, 2 MW.

	Predicted, minimum	Experiment
Gain factor, $\lambda \geq 4A$, CH_4H_20 grooved	5	4.7 ± 0.5
Vector flux of the source $\lambda > 4A$, S=300 cm^2	$0.5\ 10^{14}$ n/s/sr	0.34 ± 0.1
Scalar flux on a sample, L=18m	-	$1.8\ 10^6$ n/cm^2/s

URAM-EXPERIMENTS: INSTALLATION & IRRADIATION CONDITIONS

The basic element of the URAM installation is an Al framework (1) cooled with gaseous helium to 25-20K, Fig 2. Two cylindrical cavities (2) are cut inside it, 70mm in diameter and 10 mm thick, to condense methane. A membrane (3) forms the side of each cavity, and is welded into the framework. Its displacement is detected with induction gauges (4). Helium passes through many small channels (5) that are cut to enhance the heat transfer area. The framework is placed inside a vacuum jacket (6). The gaseous methane supply, and extraction of radiolytic hydrogen too, pass through a single access tube connected to a vacuum pump (7) and a pressure bottle (8). When the latter is valved off, gas space in the tube amounts to 2.3 ltr. Temperature of the methane can be controlled with the helium flow rate within a range of 22-60K (as measured in the centre of the solid methane bulk) and detected by two themocouples. All parameters (pressure, position of membranes, temperatures) are recorded in computer files for off-line processing.

Heat production in URAM methane due to fast neutron slowing-down was estimated to be 0.055 W/cm^3. This is based on the spectral density distribution of neutrons measured by using the threshold detectors method. Together with heat production due to γ and β radiation, this adds up to 0.06 W/cm^3 (+10%) and corresponds to the heat

production in the real CM. The calculated temperature difference in the bulk of methane is 10-12K which also corresponds to the gradient in the CM.

Seven runs of CH_4 condensation followed by irradiation were accomplished, with irradiation times ranging from 15 to 95 hours. Irradiation temperatures varied from 22 to 60K (in the centre of methane bulk); minimal temperature was 15K (in the outer area of a sample). During some runs, radiolytic hydrogen release was forced daily by heating methane up to 65-70K.

URAM-EXPERIMENTS: RESULTS & ANALYSIS

Swelling of Methane.

It was planned that a primary task of the URAM-experiments was to observe the swelling of methane due to radiation decomposition, by means of measuring the position of membranes at constant pressure and temperature conditions. However, no significant displacement of membranes that could be attributed to swelling was detected, even after 40% of the methane had been disintegrated as a result of 4-days irradiation. This is not surprising if one considers that the products of CH_4 decomposition are H-saturated hydrocarbons. Therefore, the number of molecules stays almost constant during irradiation. Moreover, it is easy to estimate that the end products occupy space of a lesser volume than the volume of source CH_4 molecules (if radiolytic hydrogen does not form bubbles!). Approximately, the ratio of volumes is (5n+1)/6n, where n is a number of carbon atoms in a daughter hydrocarbon.

Radiolytic Hydrogen: Radiation Yield.

Radiation induced formation of H_2 molecules was estimated by two methods - analysing the rate of pressure increase in the tube at equilibrium at 55K (when production of hydrogen equals to its release from a sample) and by measuring the pressure in a tank during the heating of the methane, at just the moment when hydrogen has already left the methane bulk, but methane has not yet started to vaporise.

Output of radiolytic hydrogen on the first day of each run of irradiation appeared to be about twice as much as afterwards: $(4.8 \pm 0.3)\ 10^{-3}$ mol/mol/ hour against (2.4 ± 0.2). These values are G=6 and 3., respectively, expressed in the conventional unit of "radiation yield", molecules per 100eV. The first value corresponds to the referenced

H_2 output for both gaseous and liquid phases of methane [5]. The second is 20% greater than the H_2 output of solid methane at the LINAC machine moderator in Japan [1] and about twice as much as Dr J Carpenter's value for CH_4 irradiated at 10-15K [3]. Taking into account these two referenced values and the fact that the irradiation temperature oat URAM was higher than 20K (about 25K space-averaged), we should make a conclusion regarding the tendency to H_2 output to decrease at lower temperatures. Probably, this has to do with the two phase transitions in methane at 20K and 12-14K.

Radiolytic Hydrogen: Diffusion in Methane.

One reason that could cause swelling of methane might be a formation of hydrogen bubbles but it became evident from the analysis of H_2 release that the radiolytic hydrogen exists in methane mainly in a solution. Really, a long-term monitoring of gas pressure in the access tube at 55K, 45K, 35K, 25K has showed us that hydrogen continuously releases from the matrix during irradiation; the higher the temperature the faster it passes to the access tube, see fig. 3. Diffusion is rather slow: to reach equilibrium at 55K, it takes about 2.5h. At T<30K the rate of H_2 leakage is too small to be detected. Behaviour of H_2 in the methane matrix at T<60K is consistent with a diffusion mechanism ruled by Arrhenius expression $D=Do\ \exp(T_{act}/T)$ with effective parameters $Do \simeq 0.005$ cm^2/sec and $T_{act} \simeq 300$K.

During transients, when methane is heated fast enough, hydrogen, stored in matrix at lower temperatures, releases much faster at T>55-60K - by about two orders of magnitude more than it should if diffusion is ruled by the parameters above. This process takes some minutes, see Fig. 4. During burps, H_2 leaves the methane in seconds. Such a fast diffusion is consistent with penetration of solid methane as in a gas phase, or through cracks. Calculation for a gaseous model gives $D \simeq 0.02$ cm^2/sec - just the value needed to explain the hydrogen diffusion at 65-70K. It is interesting to note, that dependence of the diffusion rate on temperature at high temperatures appears to be nearly the same as at low temperature. This was concluded after analysing the dependence on irradiation time of the H_2 fraction released due to a burp, see Fig. 5 (analysis is too difficult to be displayed in the paper). One possible explanation of such behaviour of H_2 is that hydrogen gas travels through cracks, but that the temperature dependence of diffusion is defined by H_2 molecules diffusing inside crystallines.

Radicals: Observation of "Burps" & Analysis.

As it was observed and interpreted by Dr J Carpenter [3, 6], radiation induced radicals, mainly CH_3, can be stored in methane and recombine violently in a spontaneous way or owing to a slight change in the cooling conditions. He called the occurrence "a burp", because an ejection of radiolytic H_2 usually accompanies it. In chemical kinetics, such a phenomenon is called a "thermal explosion". The critical conditions, a relationship between radical concentration, N, temperature, size of a methane slug and cooling rate, have to be fulfilled for a fast reaction of radicals recombination (RRR) can occur. A lot of expressions for the critical condition are known; one is Dr Carpenter's derived for a slug of methane of any shape, cooled from outside and heated inside, but neglecting temperature Laplacian in the heat balance equation:

$$Q_R \cdot T_{act} \cdot K(T) \cdot N^2/(\alpha \cdot T^2) = 1, \qquad (1)$$

where Q_R is an energy of recombination, T_{act} is an activation temperature, K(T) is an Arrhenius factor, $\alpha = q/(T-T_0)$, q is the heat density due to irradiation and T_0 is a coolant temperature.

Another is D A Frank-Kamenetsky's, the classic of chemical kinetics theory [7], derived for a slab slug of thickness "a", with no heat sources, except RRR, at a given periphery temperature, but with Laplacian in the heat balance equation:

$$Q_R \cdot T_{act} \cdot a \cdot K(T) \cdot N^2/(\lambda \cdot T^2) = 0.88, \qquad (2)$$

where λ is a thermal conductivity of methane. Eq. (1) can be transformed into (2), except the right term, when thermal conductivity is poor, and $1/\alpha \simeq a^2/\lambda$.

Unfortunately, none of these could be applied to our case, because the space distribution of temperature, due to the high density of energy release, was too steep to neglect. It causes the T value needed for Eq. (1) or (2) to remain unknown. Therefore, nothing could be derived except a single conclusion that "at a constant irradiation temperature T>20K the critical conditions for spontaneous development of a burp are not satisfied, however long the irradiation proceeds". This was confirmed in the URAM experiments. But, it appeared that fast RRR can be ignited by increasing helium temperature after methane had been irradiated for more than 4 hours at T<28-29K. The temperature rise needed to ignate a burp was 3-5K. Some pictures of burps are in Fig 6 and 7.

Sixteen pairs (in both chambers) of burps were detected. For two them, the maximum methane temperature reached about 90K at outlying parts of a bulk. The burp data gave us an opportunity to estimate a rate of radical recombination energy accumulation - (140 ± 20) J/mol CH_4 per hour, or (2 ± 0.4)% of an absorbed dose rate. This value is consistent with Carpenter's value. Considering that CH_3 radicals are relevant to a RRR process, the rate of their production is R≃(0.84 ± 0.12) 10^{-3}m/m per hour, or (1.3 ± 0.2) 10^{-7} mol/J - one-third as much as the H_2 production rate.

Another value that can be estimated from the burp data is an activation energy (we will use it in terms of temperature), the parameter in the Arrhenius law for RRR. By a theory, $N \alpha \exp(-T_{act}/nT)$ at saturation, where n is the power law of the reaction. Then, an energy yield in a burp, Q, and irradiation temperature T should satisfy an expression:

$$\ln Q = T_{act}/nT) + \text{const.}$$

Actually we have used an expression:

$$\ln Q_1/Q_2 = T_{act}/nT_1 \cdot T_2 \cdot (T_2 - T_1) + \text{const}, \qquad (3)$$

for pairs of burps occurred simultaneously in both methane cavities - 1 and 2, because variation of irradiation temperature in time vigorously affects the saturation concentration of radicals. It allowed us to avoid the effect of temperature variation.

Expression (3) was confirmed, see fig 8, for the burps with irradiation time more than 7h. The activation temperature value appeared to be (290 ±30)K, if one takes n=2. This value is twice J. Carpenter's T=155K estimated from two burps observed. This discrepancy is not surprising. But we derived a surprise conclusion from the analysis of the temperature behaviour at the leading edges of burps: the drastic temperature rise, up to 15K/sec could not be understood if T_{act} = 300K or 150K. Calculations based on the classical model of thermal explosion give T_{act}>700K for n=2 or even more, if n>2. There are two ways to explain this clumsy situation.

1. Different chemical reactions are responsible for fast RRR during a burp ("hot reaction") and for recombination of radicals at lower temperatures during storing ("cold reaction"). So, Arrhenius factors K_{cold} and K_{hot}, that

are consistent to these mechanisms of RRR, should satisfy the conditions: $K_{cold} > K_{hot}$ at 25K but $K_{cold} << K_{hot}$ at T>30K

2. Fast RRR has a nature other than thermal explosion, maybe, of a chain reaction.

This conclusion is, in fact, rather of a theoretical than practical interest. On the contrary, the dependence on irradiation time of H_2 fraction, released after a burp, (see in fig 5), is of importance for practice. It allowed us to decide how fast annealing of radicals should be repeated to prevent damage of the CM moderator in a casual H_2 release. Now we estimate that it has to be done 3 times a day.

Finally, some information about the rates of "cold" RRR, if it obeys a 2-power reaction:

the rate of RRR at 26K $K_{cold} = Ko \exp(-300/T) \simeq (37 \pm 8)$ mol/mol per hour;
the same at 20K $K_{cold} \simeq (1.3 \pm 0.4.)$ mol/mol per hour;

$Ko \simeq 10^6 - 10^7$ m/m/hour. For comparison, Ko for gaseous phase of CH_3 is equal to $1.7.10^3$ m/m/h.

The time constant for relaxation of the density of radicals to its equilibrium value $\tau = 5.7$ hours for 26K and 30 hours for 20K.

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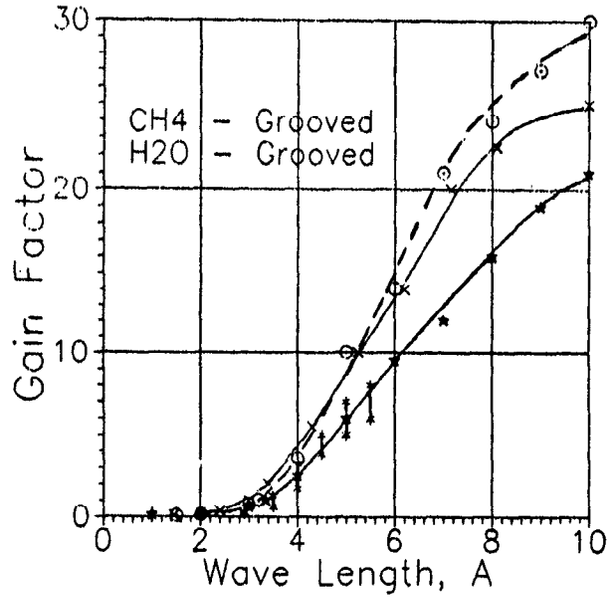
Fig. 1. Neutron flux of the CH4 cold moderator at IBR-2 compared with that of the light water, room temperature moderator (measured at four neutron beams.



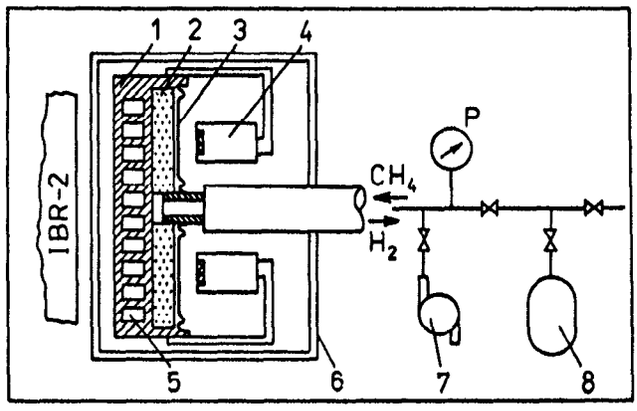


Fig. 2. The sketch of the URAM facility (for comments see the text)

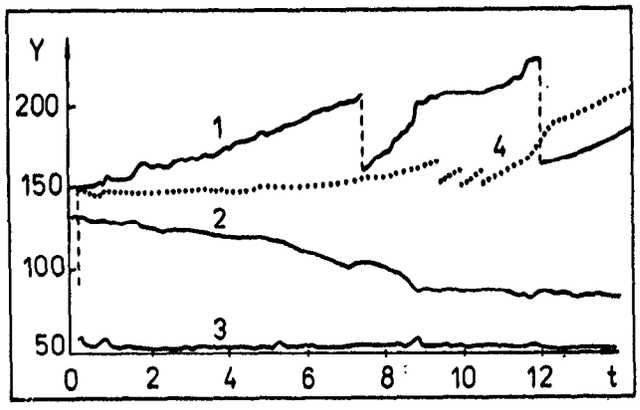


Fig. 3. The diagram of hydrogen release during irradiation.
1- pressure in the tube at t=55k; 2- displacement of the membrane; 3- methane temperature,K; 4- pressure in the tube at T=45K. Time scale: hours

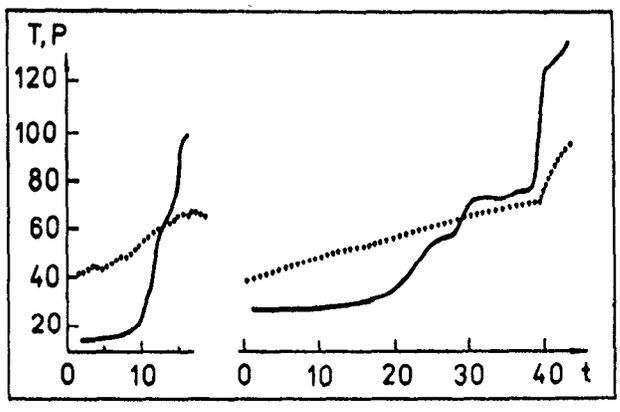


Fig. 4. The diagrams of hydrogen release during heating the methane.
Solid lines are pressure in the tube; dotted - temperature of methane.
Time scale: minutes

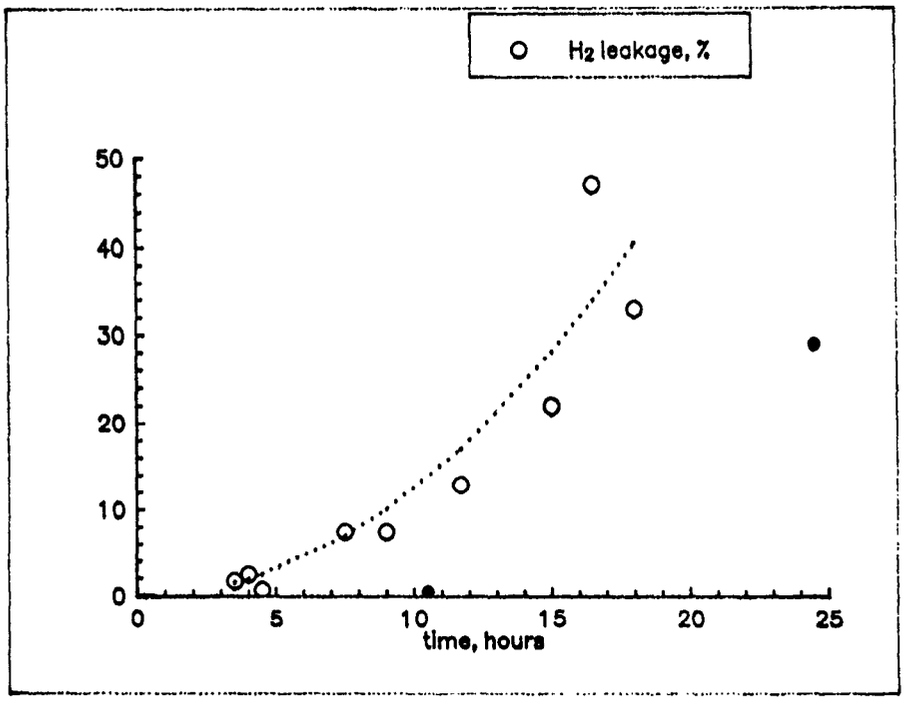


Fig. 5. Hydrogen fraction released after burps.

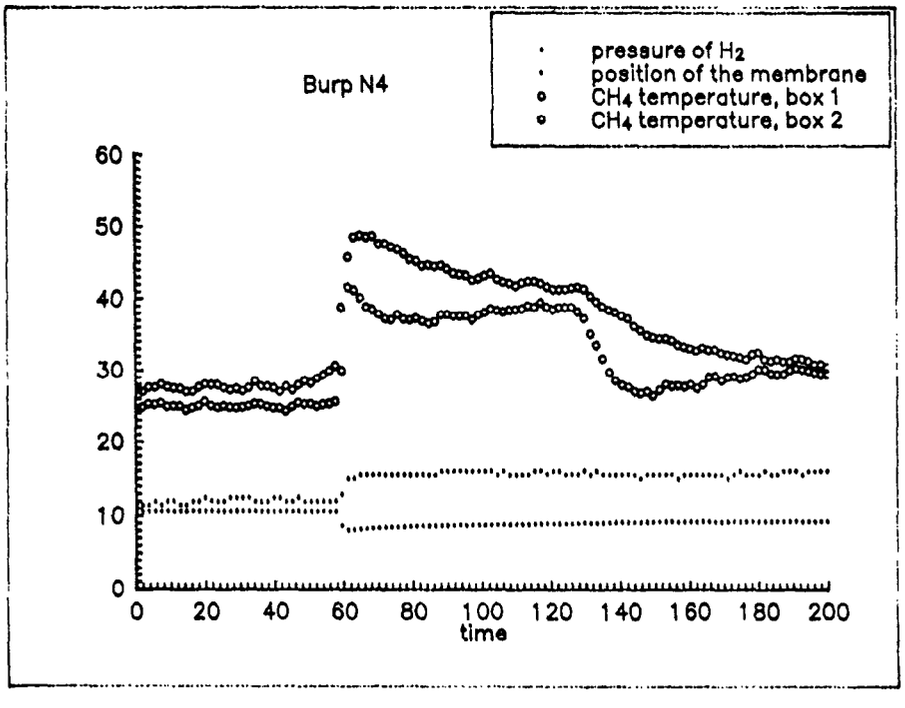


Fig. 6. Time diagram of methane parameters during the burp. Time scale is in seconds, temerature - K degress, pressure - mbars.

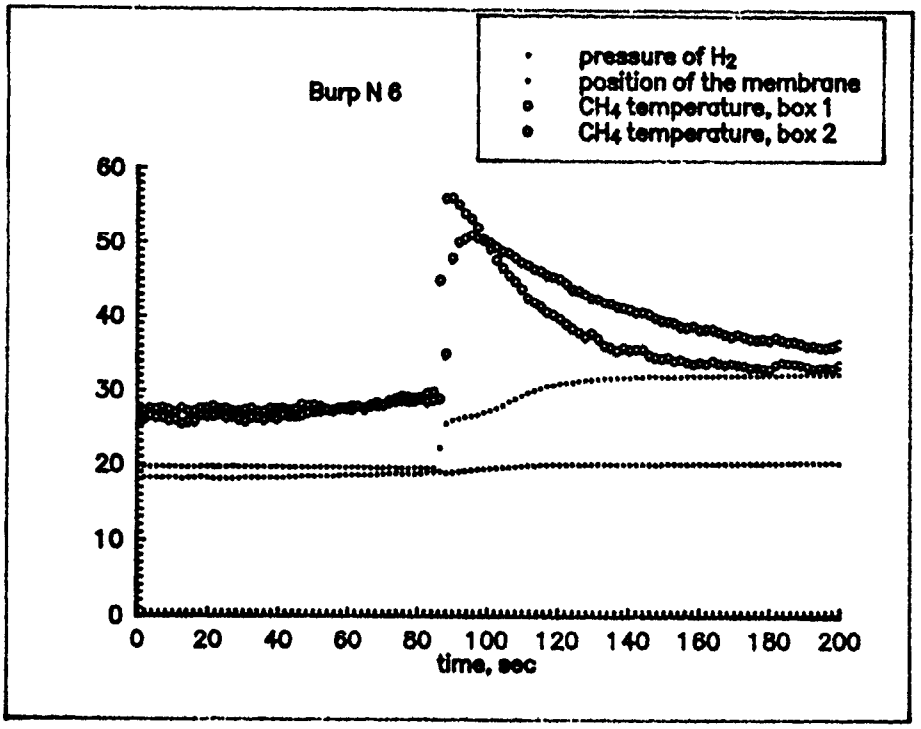


Fig.7. Time diagram of methane parameters during the burp.
Time scale is in seconds, temperature - K degrees, pressure - mbars.

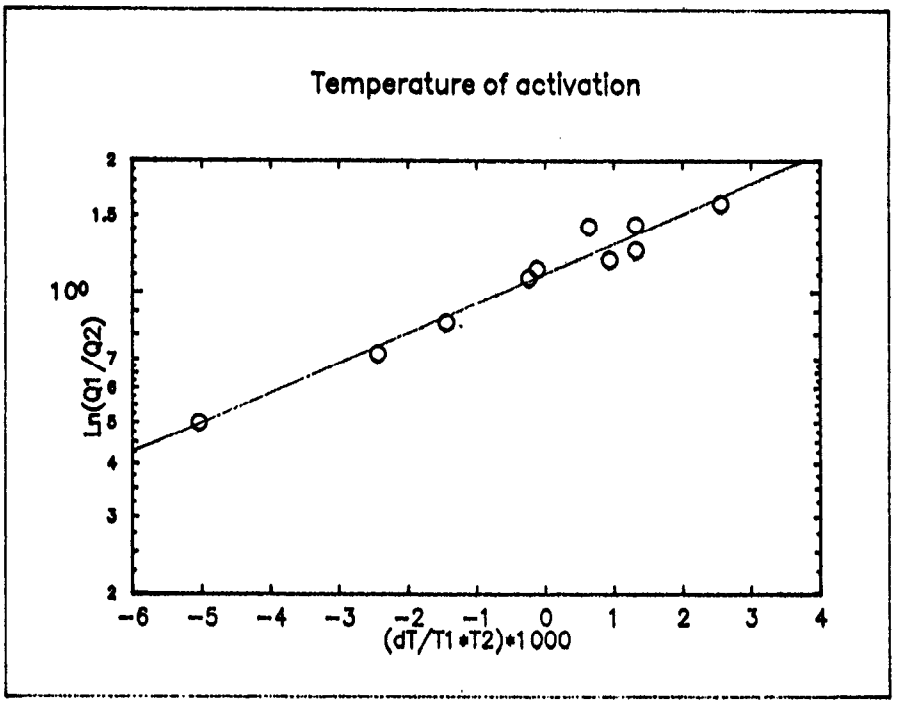


Fig. 8. The diagram for activation temperature estimation from the dependence of a burp energy on irradiation temperature (see text for more comments.