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ON RADIATION EFFECTS IN WATER ICE AT LOW TEMPERATURES

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ABSTRACT

In the paper an attempt is made to come to conclusion on possibility and conditions of fast release of stored energy in water ice irradiated with neutrons. In particular, the rough estimate of maximum allowable residence time of ice pellets in the supposed ESS moderator, before it suffers of sudden leap of temperature, is done having based on the scanty experimental data and some theoretical models of fast recombination.

1. Introduction

Recently, efforts have been started in various places to improve the performances of cold moderators in one way or the other. In particular, such combinations as of water ice and hydrogen, methane and water ice (clathrate) are considered to be effective for pulsed spallation neutron sources of the multi-megawatt range of beam power [1]. Being superior in neutronic performance to the currently used liquid or supercritical hydrogen, they would be "the hard nut to crack" because of their bad resistance to radiation.

Under radiation, free radicals are produced in many chemical compounds, and the candidates for the moderator material in question (methane clathrate, water ice, solid methane) are of no exception. At low temperatures, their accumulation gives rise to the stored chemical energy in an irradiated sample as recombination of radicals is an exothermic reaction. Usually, the process of fast recombination can be stimulated by heating of the sample but under specific condition, the stored energy may be released spontaneously, with no perturbation in cooling condition [2-10]. Naturally, sudden leap of temperature in the process has an ill effect on a yield of cold neutrons from a moderator.

This phenomenon is revealed by solid methane under fast neutron radiation in temperature range 15-25K; keeping up the tradition started by J. Carpenter, the author refer to this phenomenon as "a burp" or "burping.

Keywords: Cold moderator, radicals, stored energy

Unlike solid methane, only a little is known about burping of water ice under neutron radiation. Moreover, we lack a good deal of information about kinetics of radicals in ice which is necessary to make a conclusion on possibility and conditions of burping. Nevertheless, in the paper an attempt was made to have received at least the qualitative estimate, based on the scanty experimental data and some theoretical models of fast recombination.

As to the methane clathrate, there is no information about its radiation resistance properties. But, it is evident that radicals are expected to form faster than in pure water ice, due to higher opportunities for new radical producing reactions (such as $CH_4+H_2O^+ = CH_3+ H_3O^+$ that is exothermic with a very high cross-section) and a higher yield of H-atoms from methane. Therefore, methane clathrate seems to have to be prone to burping as well but no estimate can be made until irradiation experiment on it is performed.

2. Radiation yields of radiolysis products and kinetics of radicals

Radiation yield *G* is usually defined as the number of species per 100 eV absorbed energy of radiation. Main products of radiolysis of water ice are:

Molecules: H_2O, H_2O_2, H_2, O_2;

Radicals: H , OH, HO_2, H_3O;

Ions (intermediate products): H^-, H^+, H_2O^+, H_3O^+ etc.

Many factors affect *G*-values, such as LET (linear energy transfer), isotopes contents (heavy ice is damaged less than the light one), phase state and structure (for amorphous structure, the yields are several times higher, than for crystalline structure), impurities [11-24]. Temperature of irradiation has no effect upon the primary yield of radicals (that is, just after a track) but strongly determines their concentration in the course of irradiation.

The trapped hydrogen atoms in irradiated ice have been found to react over a large temperature range; there are stabilized atoms which react slowly at 60 K, as well as atoms which react rapidly at 15 K. No hydrogen atoms are stabilized at 77K. Decay of H-atoms shows a stepwise character instead of obeying a power law like $dH/dt = k (H)^n$; along a short temperature interval kinetics can be described in the usual way with the activation energy about 1 kcal/mol (500 K) at 30 K, and 1250 K at 40-50K, see in Fig.1 [19].

The rate of decay of H-atoms is relatively low, and at 20-30 K for short time interval (several minutes or an hour) we have final yield of H almost equal to the stabilized yield 0.33.

Following is a purely phenomenological model of the radicals behavior in cold ice by S.Siegel [20]. Highly energetic electrons formed by the interaction of primary particles with electrons in the ice matrix lose the energy in discrete bursts of approximately 60-100 eV per burst. This energy first exists as collective excitation involving the entire manifold of molecules. The collective excitation then degrades into the excitation of several H_2O molecules. In their turn H_2O molecules either degrade their extra energy into heat or dissociate to form OH radicals and H atoms. The distance between successive energy-transfer bursts from the primary electrons is of the order 4000 A°. Since diffusion is relatively slow at T<77K, it is possible to consider as independent the initial events in each energy transfer site, i.e., in each spur. If an H atom is going to recombine with an OH radical, it will do so within a relatively small volume

encompassing the point of formation. The probability of the reaction will depend only upon the number of OH radicals in the volume. OH radicals become mobile only at 110 K . Their activation energy is about 60 kcal/mol, 31000 K; the kinetics of OH is not quite clear. Anyway, OH radicals remains almost untouched to 80-90K, except some losses due to H+OH and $OH+H_2O^+$ reactions.

Radiation yields of some products of H_2O radiolysis under γ-radiation are in the Table 1.

Table 1. Yields of H_2O ice radiolysis products *G*, molecules/100 eV

Primary yields, $t < 10^{-6}$ sec *):

H	OH	H_2	H_2O_2
3.6	3.6	0.45	0.8

Stabilized yields, $10^2 > t > 10^{-6}$ sec:

0.9	0.8	0.45	0.8	T=4.2K
0.33	0.8			T=77K
(0.23)	(0.4)	(0.84)	(0.86)	water, 300K

Final yields of stabilized products, t >1 hour, low doses:

0.9	0.8			T=4.2 K
0.7-0.8	0.8			T=20 K
0.4-0.5	0.8			T=30 K
0.15-0.2				T=40 K
0.05	0.6-0.8			T=77 K
0	~0.6			T=110 K
0	~0	~0.8	~1	T>120 K

*)- *t* is the time after the primary ionization.

3. Experience on burping effects in water ice

Apart of solid methane, burping (that is, fast release of stored energy) of water ice is not yet studied except only casual experience.

3.1 Burping effects in H_2O-containing ices under UV-radiation.

A great number of experiments with mixtures of ices H_2O, CO, NH_3, CH_4 to confirm the hypothesis about the origin of organic life in grains of cosmic dust clouds have been performed by astrophisicists (Greenberg, Allamandola, Pironello, Strazzula, Schutte et al. [25-29].

Experimental condition :of the experiments:

Samples - thin layers of amorphous ice , 0.0001- 0.1 mm;

Irradiation temperature - 12 K;

Dose rates - much higher than expected at ESS .

Results of the experiments those of interest for cold moderator applications, are as follows:

- For samples of any size and composition, fast release of energy and flashes of light after warming to some temperature have been observed; for compounds with a high content of water, this temperature was 27 K.
- Energy released in water ice corresponds to 2.4 % of the saturated concentration of radicals.
- No sudden, spontaneous release of energy has been observed.

3.2 Sudden release of energy in the ice moderator of the NIST reactor.

Several years ago a cold moderator of the research reactor of the National Institute of Standards has been in operation [30-31]. Without annealing, it showed burps after 3-5 days of continuous operation with released energy >1 MJ. Conditions of irradiation and burps were as following:

- (93% heavy + 7% light) water ice moderator of 16 litres volume;

- dose rate 37.5 mW/g;

- temperature of irradiation 30-50K;

- excess of temperature at a burp 70K, released energy 60J/g;

- sudden releases of energy were initiated by any small perturbation, either an increase or decrease of temperature;

- most probably, it was OH+H reaction which took place during burps. It gives critical concentration of H-atoms 0.2-0.3%.

4. Analysis of the experience with irradiation of ice.

Making correlation between radiation effects in water ice and solid methane, one can see close similarity in burping. Really, in both cases there is an upper threshold of temperature above which no spontaneous burp can occur; it is 25K for methane, and about 50K for water ice. Moreover, there is a lower limit of temperature for spontaneous burping in both cases as well, not yet rigorously defined till now. It is somewhere between 10K and 15K for methane, and 27K, or a little bit less, for ice. There below, we are trying to analyze the situation.

First statement I'd like to stress at, is that , like the case of methane discussed in [32], the process of uncontrollable release of energy in water ice is properly a propagation of the front of recombination through a sample or propagation of soliton, not thermal instability of the slug irradiated. Critical condition for a burp is not governed by Frank-Kamenetski's equation as it is often referred to [33]:

$$\frac{Q \cdot T_{act} \cdot x^2 \cdot K(T) \cdot n^2}{\lambda \cdot T^2} = 0.88, \qquad (1),$$

where λ is the thermal conductivity, Q is the molar heat of recombination, $K(T)$ is the factor of recombination rate at temperature T usually obeying Arrhenius law with "activation energy" T_{act}. That can be proved by an analysis of the experiments with thin films of ice, mentioned above. Indeed, applying Eq.(1) with x (thickness of the film) equals to 0.01 cm (the largest value in the experiments) and with other values more or less defined, we receive $K(27K) \approx 0.1\text{-}1.0\ c^{-1}$. With such large value of the factor of recombination rate, it is impossible to understand why the saturated concentration of radicals at 40 K - 50 K amounts to 0.4-0.6% as it was in NIST case.

The same one can conclude calculating Eq.(1) for Greenberg's and NIST cases, i.e., burps at 27K and 40K. Parameters x and $K(T)$ in the NIST case are both 2-3 orders of magnitude greater, whereas other parameters in Eq.(1) are almost equal for both cases. So that, at least for one of two cases Eq.(1) does not work.

The weak dependence of critical concentration upon both temperature and a size of irradiated sample proves the conception of burping as a propagation of the front of recombination through the sample after the process was ignited at one small region of appropriate size. Moreover, we are urged to take into account the effect of nonuniformity of radicals on the critical concentration value as well. For the followiing consideration, let us use the critical conditions derived in Appendix A:

Being based on the conception of a burp as a propagation of recombination front, one seemingly jumps to the conclusion that spontaneous release of stored energy takes place elsewhere below some temperature at which critical density of radicals is equal to the saturated value of radical concentration. Indeed, the latter is the fast dropping function of temperature (see Appendix B) whereas the former is almost temperature independent in some temperature range, Eqs.(5) and (5a), see Fig.2. But in fact, at very low temperatures a spontaneous release of energy was observed neither for solid methane, nor for water ice. This phenomenon is still to have to be understood. Two options are open for discussion:

- 'Hot Point' version: at very low temperatures an irradiated sample is kept supercritical, instable, but probability for the rise of a 'hot point', the initiator of the chain recombination process, is too small for the period of expectation time. This explanation seems to be realistic as, regardless of the nature of hot points, their appearence should be strong temperature controlled.
- 'Crack Recombination'-version: if consider mechanical model of recombination, the critical concentration would have steep character at low temperatures, see (5a) in Appendix A. This could explain the existence of the lower limit of the critical density (probably, 27 K for water ice), as easily as it was done with the hypothesis of "hot point". Thus, in (5a) $n_{crit} \to \infty$ when $\delta \to \chi$.

There is no way to choose between two hypothesis except by experimental investigations.

5. Estimation of radiation effects in ESS moderator materials (burps)

Experience with irradiation of cold ices, together with up-to-date understanding of burp mechanism, enable us but roughly and not reliably, to have predicted possible radiation effects in water ice and solid methane subjected neutron radiation of ESS conditions which are:

- temperature of irradiation: 20-100K;
- dose rate : 1.3 W/g (water), 2 W/g (methane);
- samples: a batch of beads, 2-3 mm each, in contact with neighbours and with coolant.

Let's start consideration from low irradiation temperature 20-25K. In accordance with [19], G_H is about 0.7 at 20 K, if we take into account recombination of H atoms in a short time, about half an hour after formation. So that, rate of H-atoms production is 4-5 times faster than for NIST, at a given dose rate. Accounting for the real dose rate, we have radical production at ESS ice moderator equal to ≈ 140 of that of NIST, i.e. about 0.7% per hour. The critical concentration by HP version of ignition of a burp, Eq. (5), would be less than 0.6%, because the critical concentration in the case of neutron radiation is definitely lower than for NIST, where the most part of radiation was due to gamma. It means that a critical condition would be reached within one hour. It is very probable that occurrence of the hot points would be simplified by friction of pellets with each other and with walls of the chamber, in contrast to the case of Greenberg's experiments. So that, burping in ice pellets might be expected to occur faster than each hour at irradiation temperature 20K-25K.

If CC-version of criticality condition is true, then no burp occurs, no matter how long samples were being irradiated. Actually, saturated concentration does not depend on dose rate but temperature only (see Eq.6a in Appendix B), and therefore, would be less than 2.4%, Greenberg's value for 12 K, whereas the critical one would be higher, see Fig.2.

As to Table I, production rate of radicals at 40 K is 4-5 times lower which gives more time for building up of critical concentration, that is about 3 hours. Thus, we have a satisfactory data on burps at higher temperatures, but information is insufficient for lower temperatures <30 K. Also, it seems important to know whether friction of pellets takes an effect on initiation of a burp.

A rough notion on temperature dependence of the burp expectation time is given by Fig.3. The functions in the plot were calculated with the equation:

$$\tau_{res} \approx -\frac{n_{sat} \cdot ln(1 - n_{crit} / n_{sat})}{\dot{R}} \qquad (2),$$

which is based upon the time dependence of radical concentration given by Eq.(6):

$$n(t) = n_{sat} \cdot (1 - exp(-\dot{R} \cdot t / n_{sat}) \qquad (3),$$

where $R=1.3 \cdot D \cdot G$ is the radical production rate, mol.%/hour , D is the dose rate, W/g, and G is the radiation yield of radicals. In the case of water ice, the radical production rate R strongly depends upon temperature, see Table 1. The critical concentrations were calculated with Eq.(5a), with parameters of the equation satisfying two experimental points available and assuming the $T^{1/2}$-dependence of δ upon temperature.

For the solid methane, situation is quite similar except for more properly defined the temperature limits.

6. Summary

It was shown that water ice expected to display the same effect of burping under radiation at low temperature as solid methane does. Differences would be in a temperature range where the phenomenon takes place, and in a quantity of absorbed dose needed to cause it.

7. Acknowledgements

The author is very thankful to Dr. Guenter Bauer for formulation of the problem, and to Dr. E. Roumanov who has granted the result of his work not published yet.

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Appendix *A*

Following is the simple deduction of a critical condition for propagation of recombination process through a bulk. Assumed approximations are:

- radicals are distributed in a clustering way with a density of clusters being uniform, and with a concentration in a cluster n_{cl} much more than averaged concentration n;
- all radicals in a cluster recombine quickly ('quickly' means a span of time less than r^2/χ, where r_0 is the size of a cluster, and χ is the temperature conductivity coefficient) if temperature exceeds some threshold value T_{cl}.

Propagation of recombination is provided if burning of one cluster gives rise to recombinations of at least one adjacent cluster. With the assumptions given above, it gives three equations to be solved:

$$(n/n_{cl})\cdot(R/r_0)^3 = 1 \qquad (4.1)$$

(this says that inside R around the burned cluster there is one adjacent cluster),

$$(T_{cl} - T_0) = T(R,t) \qquad (4.2),$$

$$\partial T(R,t)/\partial t = 0 \qquad (4.3).$$

Eqs. (4.2) and (4.3) follow from a condition that transient temperature $T(R,t)+T_0$ reaches T_{cl} just at the distance R from the centre of the burned cluster. Transient temperature function was used in the form of "prescribed diffusion":

$$T(r,t) = \frac{Q n_{cl} e r_0^3}{c_p \sqrt{(4\chi t + r_0^2)^3}} exp(-r^2/(4\chi t + r_0^2))$$

where Q is the molar heat of recombination, c_p is the molar heat capacity.

After calculations we have from (4.2) and (4.3):

$$R = r_0 \cdot \sqrt[3]{\frac{Q n_{cl}}{c_p (T_{cl} - T_0)}} \qquad (4.4).$$

Finally, from (4.1) and (4.4) a fairly evident equation follows:

$$n_{crit} = \frac{2 \cdot c_p \cdot (T_{cl} - T_0)}{Q} \qquad (5).$$

This value of the critical concentration is actually the upper estimate, as clusters have a finite size, and may be separated more distantly from one another to catch up the same temperature.

The rigorous critical condition seemingly needs the terms of both the temperature conductivity coefficient and the reaction rate at T_{cl} to account for the finite time of recombination inside a cluster. But it can be shown that the condition (5) is sufficient provided the origin statement of the "flammabillity" of a cluster at the temperature T_{cl} .

The latter means that the condition $r^2 > 4\chi\tau$ should be fulfilled, where τ is the time of recombination at T_{cl} .

Another probable model of fast recombination is thought to be one that accounts for acceleration of diffusion of radicals in cracks caused by a transient temperature gradient.

A theory of fast recombination, accounting for both thermal and mechanical nature of recombination, is currently under derivation by E. Roumanov (Institute of Macrokinetics, Moscow). This model seems to be the most applicable to our case. The expression for the critical concentration of radicals appeares to be in the form

$$n_{crit} = \frac{c_p \cdot const}{Q \cdot (1 - \chi / \delta)}. \qquad (5a)$$

instead of (5), where δ is a value characterizing the extent to which crack formation affects the recombination process. By physical reasons, it should slightly depend on temperature being increased with a rise of temperature.

Appendix *B*

As it was shown in [32], concentration of radicals during irradiation is ruled by the equation:

$$\partial n/\partial t = R - K(T)\cdot n^2 - R\cdot C_1 K(T^*)\cdot n \qquad (6),$$

where R is the rate of production of radicals due to ionizing radiation, $K(T)$ is the factor of recombination rate at temperature T_0 (usually, Arrhenius factor $exp\,(-T_{act}/T)$, and T^* is some effective temperature at which the radicals recombine near a track, not a transient one, $T^* \approx \Delta T + T_0$, where T_0 is the bulk temperature. For high dose rate, the third term could be greater than the second term, so as

$$n_{sat} = C_1^{-1} \cdot K^{-1}(T^*)\cdot \qquad (6a).$$

The dependence n_{sat} upon irradiation temperature T_0 has a steep character. However, at very low temperatures below 20 K, the relations (6a) would give too high concentration exceeding density of radicals inside tracks of ionizing particles. It seems unreasonable. Therefore, Eq. (6) fails to predict the real density for low temperatures, and it would be justified to consider the saturated density being temperature independent at low irradiation temperatures.

Finally, qualititave temperature dependence of the saturated density of radicals together with critical density is given in Fig.2.

ICANS-XIV ***The Fourteenth Meeting of the International Collaboration on Advanced Neutron Sources***

April 30, 1999

Dear ICANS XIV Conference Attendee,

Enclosed please find three figures from Dr. Eugene Shabalin's paper titled "On Radiation Effects In Water Ice At Low Temperatures" that were omitted from the ICANS XIV Conference Proceedings by mistake. These pages have been numbered 506-a, 506-b and 506-c and are to be inserted in Volume II after page 506.

Sincerely,

J. M. Carpenter
ICANS XIV Conference Chair
Argonne National Laboratory

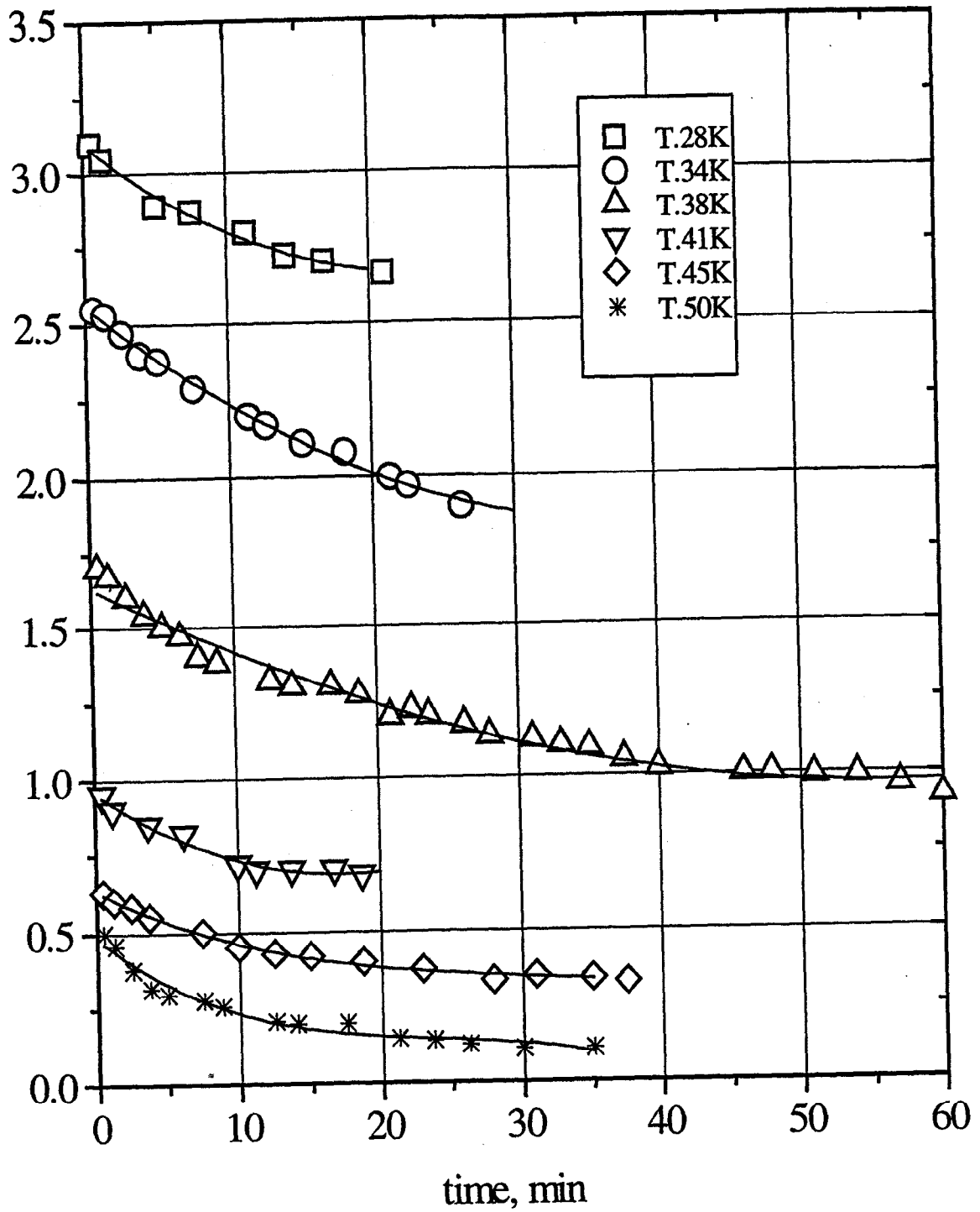

Figure 1. Disappearance of trapped H-atoms (relative concentration) in gamma-irradiated ice under isothermal conditions (see the legend for temperature); the time axis has arbitrarily been shifted to zero at the start of each isothermal measurement.

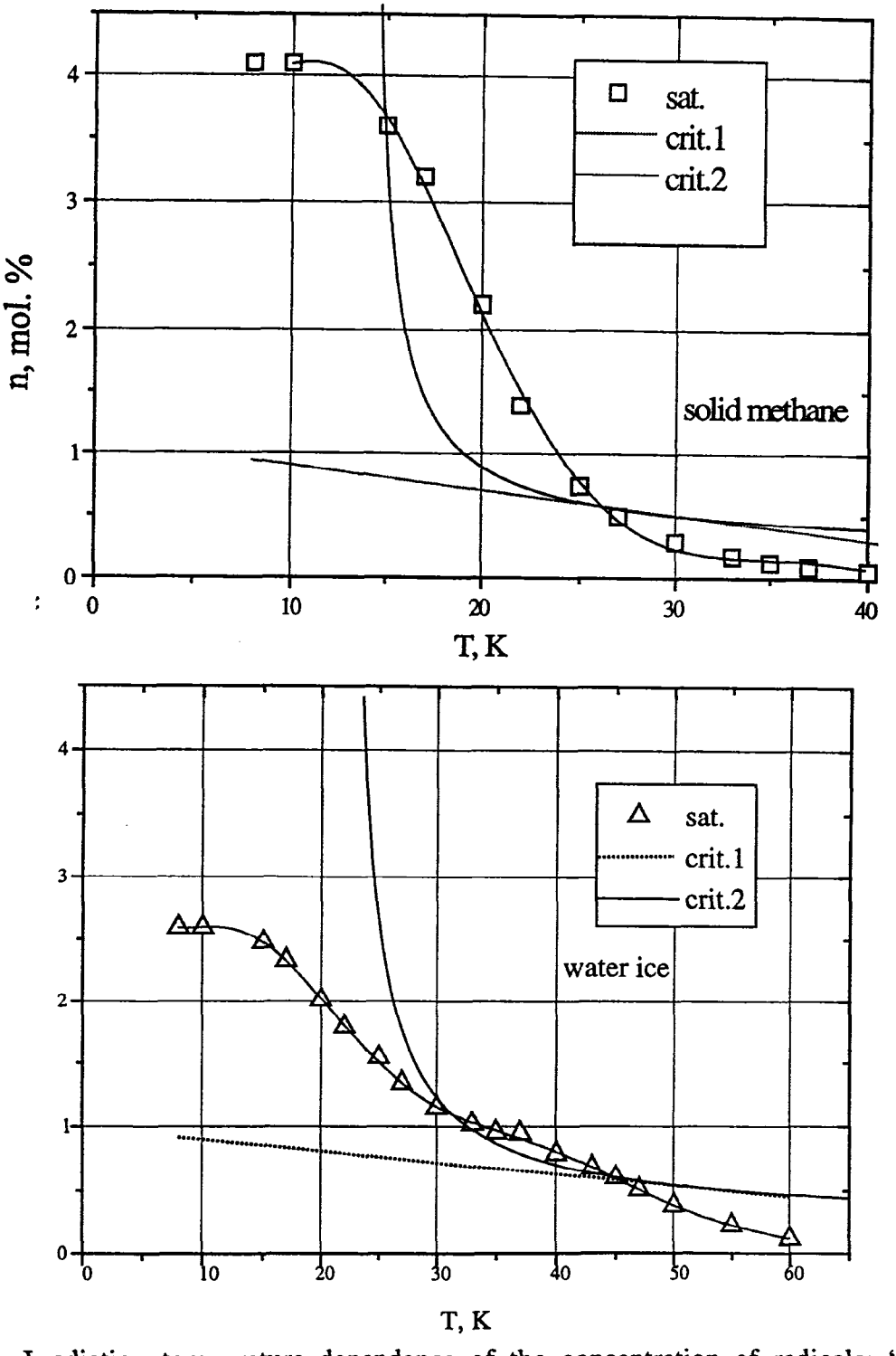

Figure 2. Irradiation temperature dependence of the concentration of radicals; "*sat*" is the saturated concentration, "*crit.*1" is the critical one to burping by Eq. (5), "*crit.* 2" is the critical one to burping by Eq. (5a). All numbers are calculated basing upon a few experimental points.

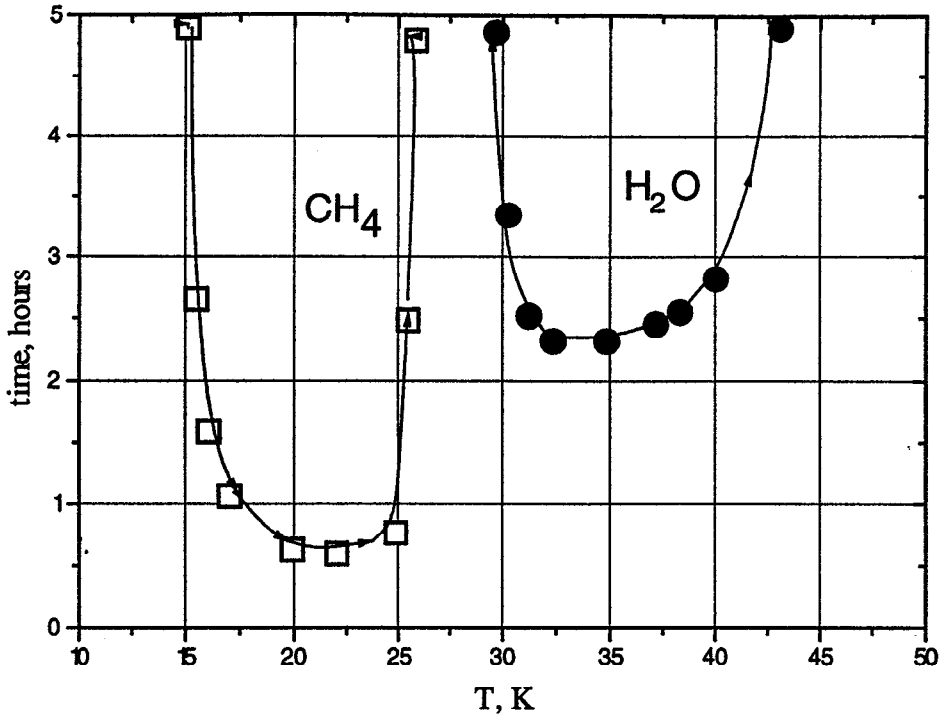

Figure 3. Estimated maximum allowable residence time of pellets in the ESS moderator versus irradiation temperature for solid methane (squares) and water ice (circles).