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TRITIUM CHEMISTRY IN FISSION AND FUSION REACTORS

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TRITIUM CHEMISTRY IN FISSION AND FUSION REACTORS

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INTRODUCTION

We are interested in the behaviour of tritium inside the solids where it is generated both in the case of fission nuclear reactor fuel elements, and in that of blankets of future fusion reactor.

In the first case it is desirable to be able to predict whether tritium will be found in the hulls or in the uranium oxide, and under what chemical form, in order to take appropriate steps for its removal in reprocessing plants.

In fusion reactors breeding large amounts of tritium and burning it in the plasma should be accomplished in as short a cycle as possible in order to limit inventories that are associated with huge activities. Mastering the chemistry of every step is therefore essential.

Amounts generated are not of the same order of magnitude in the two cases studied. Ternary fissions produce about  $66 \cdot 10^{13}$  Bq (18 000 Ci) per year of tritium in a 1 000 MWe fission generator, i.e., about  $1.8 \cdot 10^{10}$  Bq (0.5 Ci) per day per ton of fuel.

It is estimated that a 1 000 MWe fusion reactor could burn 450 g of tritium per day in a 450 ton blanket. The latter should thus produce a total  $1.7 \cdot 10^{17}$  Bq ( $4.5 \cdot 10^6$  Ci) per day, i.e.  $3.7 \cdot 10^{14}$  or  $10^4$  Ci per ton.

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However as this tritium has to be recovered on a day to day basis the concentration reached is only about 20 times larger than that which prevails in fission fuels when shipped to reprocessing plants.

#### A - TRITIUM CHEMISTRY IN P.W.R. FUELS

The fate of this tritium has been studied for many years for practical reasons. When stainless steel claddings were used a large part of the tritium found its way into the coolant. When zircaloy claddings were introduced diffusion became negligible. But, as mentioned, tritium that remains in the fuel itself ( $UO_2$ ) and that which is trapped inside the hulls end up in different parts of reprocessing plants, and in different fluid streams also according to its soluble or non soluble form.

Previous studies have shown that the fraction retained in uranium oxide is a decreasing function of the temperature of the pin cores, a rough equipartition between  $UO_2$  and zircaloy being reached at  $1000^\circ C$  - Fig 1 - from (1).

No significant amount was ever found remaining in the free space between  $UO_2$  pellets and cladding.

#### 1 - MATERIAL STUDIED

We concentrate on three pins (A 10 B, I 15 H, O 6 H) from the BORSELLE reactor in the Netherlands.

Their  $UO_2$  weight is about 1500 g (1487). They are 2975 mm long and 10.75 mm in diameter, clad with zircaloy 4,0.65 mm thick. Initial  $^{235}U$  content is 3.1%, burn-up  $32,540 \text{ MWdt}^{-1}$  at unloading.

The zircaloy to uranium ratio is .26 by weight.

In a typical pin at the time of removal an average  $2.85 \cdot 10^{10}$  Bq of  $T_2$  gas are present (0.77 Ci), corresponding to 0.3 ml NTP.

From BORSELLE three pins located on three outer rows of one bundle (a 15 x 15 type) were selected (fig. 2). During their stay in reactors bundles are moved and reoriented in order that all bundles and all pins should approximately undergo the same burn-up.

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## 2 - RESULTS

2-1 . The results of tritium analysis on three half pins are given in table I along with the percentage of tritium in the cladding. Analysis performed when dissolving the fuel prove that 99.5% of the tritium in the fuel is under soluble form (water). The scatter of results of table I between elements belonging to the same family is surprising. The center temperature of pins being about 1000°C, a 50 % average repartition was expected, but corresponds only approximately with the mean of tabulated values.

This scattered and seemingly random behaviour of tritium distribution between different chemical species is the more surprising as there is a generally satisfactory and even correlation between total tritium activity and burn-up i.e. also with location along the fuel element -fig.(3) from (2).

### 2-2 . Nature of tritiated species in the cladding :

a) Methane appears in widely varying amounts. Its formation may be either due to direct reaction of tritium with carbon dissolved in zirconium, or to the reaction of water with zirconium carbide. We would tend to favor this second route because it is a more classical reaction than the direct synthesis of methane even though, under radiation, with hot atoms, the latter has been observed (and though the amount of  $\text{CH}_3\text{T}$  observed in A 10 B, I 15 H is roughly that of T recoiling into the cladding). Also results of table II show a relative increase in  $\text{CH}_3\text{T}$  with the increase of the water form of tritium.

b) Water was found in greater quantities when the amount of tritium in zircaloy was low.

## 3 - DISCUSSION

3-1 . Tritium retained in the fuel is essentially under soluble form, and tritium in the hulls is mainly found as gas (tables III and IV). Possibly all the tritium cannot be oxidised within  $\text{UO}_2$ , as  $\text{UO}_2$  undergoing fission becomes oxygen deficient, because of the greater oxygen demand of fission products with respect to that of U itself. Non oxidised tritium would thus migrate to the cooler surface by diffusion or percolation and dissolve in zirconium.

3-2 . Differences in behaviour between individual pins could be due to differences in impurities in gases filling the free volumes inside the cladding.

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Residual oxygen or water could cause one or more of the following reactions :  
oxidation of gaseous tritium, isotopic exchange with water, oxidation of zirconium.

Either of these would make permeation of tritium into zirconium much more difficult.

3-3 . The free space inside the cladding of one pin is about 17 cm<sup>3</sup>. It is initially filled with He at one to two MPa, which means that tritium, if totally released in this volume would represent about 1000 ppm of the gas volume.

But also it only represents 50 ppb in weight of the uranium oxide (or less than 200 ppb as T<sub>2</sub>O). Which means that minute amounts of water remaining in UO<sub>2</sub>, though well within specifications (that are in the 10 ppm range), plus impurities below the 1000 ppm level in helium could explain those anomalies.

3-4 . A dark brownish color has indeed been observed on hulls from fuel elements fig (4), characteristic of zirconium oxide, and tritium could not be extracted from them by heating, before this brownish coloration had disappeared.

#### 4 - CONCLUSION

When core temperatures are higher, it is clear that a larger gradient being established tritium may diffuse out faster and escape recombining with UO<sub>2</sub>.

Formation of zirconium oxide due to varying amounts of impurities in UO<sub>2</sub> or helium may explain differences in tritium content between hulls from different pins. Variations observed between hulls from the same pin are more surprising at first.

However if in pin 0 6 H corrosion has taken place, as suggested by the methane formation and by both the low tritium content in zircaloy and the water form under which it is found, it is not a far fetched hypothesis that it did not start or grow evenly all along the pin. Many reasons, among which temperature variations, differences in impurities concentration and therefore evolution rates, would cause such differences (table V).

And the rather good correlation observed even on pin 06 between the amount of tritium recovered in the hulls and the burn-up of the adjacent UO<sub>2</sub> would confirm that local conditions govern tritium implantation in zircaloy, fig (3b).

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Observations reported here justify the practice of standardised fabrication procedures, and very strict quality controls of all materials (fuel, cladding and gases) of pins in order to lessen individual deviations of tritium inventories in each component from expected average values.

## B - TRITIUM RECOVERY FROM SOLID BREEDERS : THE CASE OF LITHIUM ALUMINATE

In contradistinction with the preceeding section measurements presented here were carried out on small samples weighing only one or two grams. Not only tritium concentrations but total amounts were therefore very small and special care has to be taken in order to obtain reliable results. Let us recall first that tritium bred in a solid blanket would be recovered by flushing it with helium gas.

### Tritium recovery experiments

Our first experiments (2) had the aim of ascertaining the nature of tritiated species extracted, as this nature governs the recovery process from the purge gas. The main effort was put on finding out what chemical modifications may be introduced by reactions with walls, or impurities in the gases, that could lead to erroneous conclusions, and to investigate the influence of texture on extraction rates.

It is established by many experiments that the main product is water, though a non negligible fraction of the tritium remains as gas (3) (4). Relative amounts of those two species however are neither fixed nor easily evaluated quantities. A consequence is that, for practical purposes, hydrogen will have to be added to the sweep gas both to drive the major part of tritium into a non condensible form and, more important yet, to limit the size of inventories (5). Diluting gaseous tritium with hydrogen also limits permeation.

In this paper we are going to describe several experiments carried out in order to establish the effect on observed species and on extraction rates from a sample of  $\gamma$   $\text{LiAlO}_2$ , of sweep rates, nature of the purge gas, etc... We compare results with observations on samples of other ceramics and try to make an interim evaluation of the situation.

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## 1 - Experimental conditions

The sample was irradiated in the previously described CHOUCA rig (fig 5) in SILOE (6). It consisted of a stack of pellets (\*) 7.8 mm in diameter, and 80 mm in height, of mean pore radius 0.035  $\mu\text{m}$ , mean grain size .36  $\mu\text{m}$  and 0.225 porosity.

It was exposed to an average thermal flux of  $1.3 \times 10^{13} \text{ n.cm}^{-2} \text{ s}^{-1}$  producing  $6.1 \times 10^{12}$  reactions per  $\text{cm}^3$  per second, or about  $3.7 \times 10^9$  Bq per day of tritium. Heating due to the  ${}^6\text{Li} (n, \alpha) \text{T}$  reaction was  $4.74 \text{ W cm}^{-3}$ , due to  $\gamma$  heating  $1.5 \text{ W cm}^{-3}$ .

The sweep gas was helium of impurity levels in vppm :

- < 0.8 for  $\text{H}_2$
- < 0.7 for  $\text{H}_2\text{O}$
- < 0.6 for  $\text{N}_2$
- < 0.15 for  $\text{O}_2$
- < 0.01 for  $\text{CH}_4$

It could be further purified by a 5 A molecular sieve bed cooled to liquid nitrogen temperature.

It could be replaced by helium-hydrogen or helium-oxygen mixtures having a somewhat larger tolerance on impurities (for instance  $\text{H}_2\text{O} \leq 2$  vppm).

The helium flow rate was usually about  $2.4 \text{ liters h}^{-1}$  the pressure above the sample  $1.5 \times 10^5 \text{ Pa}$ , the tritiated species partial pressure in the steady state 0.1 Pa.

The sample was dried in the rig, before starting irradiation, by flowing further purified helium for 48 hours over the sample heated to  $500^\circ\text{C}$ . Thirty five runs were then carried out over a period of three months (November 85 to February 86). Conditions have been described (7) (8) and will be detailed in a further report (9). We call here attention to principal results.

## 2 - Study of the rate determining step in tritium extraction when the sweep gas is hydrogen free.

When this step is diffusion in a grain of radius  $r$  the inventory  $I$  varies like  $\frac{r^2}{D}$ .  $D$  is the diffusion coefficient (10). One can thus calculate  $D$  from curves

(\*) one 30 mm, two 8 mm, seven 5 mm ones. .../

giving the tritium content in the gas phase from start-up I is just the "missing" tritium before steady state, fig (6).  $r$  is measured beforehand.

A change in temperature modifies  $D$  and induces a temporary change in the tritium content of the gas and the inventory of the solid.

The shape of the response curve should both provide the new value  $D'$  of  $D$ , and show a time dependant return to equilibrium conforming to  $D'$ . The dashed curve on fig (7) shows that this behaviour is only approximately followed, which is in agreement with many previous observations (11) : diffusion coefficients enable to predict the trend of changes in extraction rates from  $\text{LiAlO}_2$  with grain sizes or temperature, but such predictions fail to be quantitative.

The hypothesis that the rate determining step is an interface resistance transfer process depending on the difference between concentrations in the solid state and gas phases leads to an inventory law that is not easy to verify. But applying this law, in the same manner as above, provides the dotted curve on fig (8) that fits closer to experimental values. It remains to verify that this improvement in the description of results persists when applied to samples of different grain sizes, and in that case to reach a precise physical interpretation. This work is underway.

## 2) Study of the extraction rates by purge gases with 0.1% $\text{H}_2$ or $\text{D}_2$ added.

Several experiments in the literature have been reported, describing the increase in extraction rates produced by adding  $\text{H}_2$  (or  $\text{D}_2$ ) to helium or argon used as purge gas. Fig (9) displays this effect in an out of pile extraction run. It has been shown that the action of  $\text{H}_2$  takes places within the solid (2).

Assuming an interface rate determining step might provide a better explanation for this increase than diffusion limited steps, if one accepts that hydrogen reaches grain surfaces by intergranular diffusion much more rapidly than the bulk of the solid, and that it would there either displace adsorbed species or exchange with them. Such reactions are known to be rapid under favourable circumstances.

The rate at which steady concentrations are reestablished in the gas phase after a change in flow rate, compared with the time needed after modifications of either flux or temperature, also favors such mechanisms. Let us reduce the sweep gas flow rate by a factor of two, concentration in the gas becomes  $C'_g = 2C_g$ .

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As  $C_g$  is very small compared to  $C_s$ , concentration in the solid doubling  $C_g$  will be compensated by a very small relative increase in  $C_s$  in order that equilibrium be reestablished, i.e.

$$C'_s - 2C_g = C_s - C_g \quad \text{or} \quad C'_s - C_s = C_g$$

The change in average concentration in the bulk of the grain, that varies proportionally to  $C_s$  is also very small. But when fluxes are changed by factors of 1.5 or 2 inventories in the bulk of the grain change in the same ratio and response times to reestablish steady state are necessarily larger than in the previous case as shown qualitatively on fig (10) and (11). Modifying the irradiation temperatures changes release rates substantially as can be read from fig (12) and inventories vary accordingly.

### 3) Extraction by purge gases with 0.1 % $O_2$ added.

Oxygen addition to the purge gas practically suppresses the observed release of tritium.

This shows that in addition to balance considerations discussed above physicochemical reactions, probably at the surface of grains, play an important role in release mechanisms.

### 4) Comparison between extraction rates from different ceramics.

A more detailed knowledge of mechanisms could also provide explanations for the wide differences observed between effects of the parameters mentioned above on tritium extraction rates from different ceramics.

T. KURASAWA and al. have compared  $Li_2O$  and  $\gamma LiAlO_2$  (12). Aluminate displays a behaviour generally in line with the one described here, except that the authors using helium with 1000 vppm  $D_2$  seem to be able to fit release rates well to purely diffusional mechanisms fig (13). Also a temperature drop from 802°C to 557°C brings practically the tritium release to zero, during a run lasting nearly 20 hours, fig. (14 and 15). On the contrary lithium oxide releases tritium at rates insensitive to temperature variations, which indicates no retention of tritium, in the  $Li_2O$  spheres even at 470°C, fig (15) and, of course, prevents calculations of diffusion coefficients based on changes in release rates associated with temperature changes).

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In apparent discrepancy with the findings of the preceding authors H. KWAST and coworkers state that, when extracting tritium by helium (13) with 1000 vppm of  $H_2$ , even at  $460^\circ C$  the release rate establishes itself at its nominal value within a day or some hours (\*). At nearly the same temperature,  $440^\circ C$ ,  $Li_2O$  irradiated simultaneously in the same rig releases only 51 % of the calculated tritium production rate even after 25 days. That estimation of birth rates should not be incriminated seems proven by the fact that another sample of  $Li_2O$  irradiated at  $660^\circ C$  delivers its tritium at the predicted level, within apparent error limits, fig (16) and (17).

#### 5) Comparison between inventories

Another interesting comparison between tritium extractions from samples irradiated simultaneously is given in the paper by WERLE and al (8). In the same CHOUCA rig in SILOE the aluminate sample was irradiated along with four metasilicates ( $Li_2SiO_3$ ) and one orthosilicate ( $Li_4SiO_4$ ) samples.

Results respective to the orthosilicate and the aluminate are summarized in table (6).

The striking fact is that release from  $Li_4SiO_4$  is much faster than from  $\gamma LiAlO_2$  as comparable residence times are found with grain radii nearly one hundred times larger in the case of orthosilicate. But at comparable production rates, inventories that are larger in aluminate at low temperatures become smaller at  $630^\circ C$  (because of a bigger activation energy of the diffusion coefficient ?)

The study of inventories has probably to be pushed further to understand why, as has been reported, (14) and (15), in many cases the percentage of retained helium is smaller than that of retained tritium (\*\*). Recent work by BONAL (16) and fig (20) also that a step release of helium indicates at first total extraction from an aluminate sample at  $700^\circ C$ . The same sample released tritium totally closer to  $1000^\circ C$ .

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(\*) After the reactor reaches full power.

(\*\*) See fig (18) and (19) from (14).

## CONCLUSION

The behaviour of  $\gamma$  lithium aluminate with respect to tritium release has been shown to be influenced by two main factors, in addition to an obvious temperature effect.

a) a textural one : grain size. The smaller the latter the greater the extraction rates. At this stage it is not possible to decide whether this size effect can be assigned purely to bulk or surface phenomena.

b) a chemical one : the nature of the sweep gas.

Hydrogen or oxygen additions modify drastically tritium release rates through reactions taking place in the solid. Whether hydrogen acts through an isotope exchange mechanism or a reducing one or both has yet to be established.

Much less clear are reasons for differences between results obtained with  $\gamma$   $\text{LiAlO}_2$  and other ceramics irradiated simultaneously. Are the much larger extraction rates, obtained with lithium oxide, or lithium orthosilicates, due to differences in structure, are the latter the cause of differences in activation energies ? These problems are now under study.

But we can not conclude without recalling that, though tritium extraction rates are of major importance in evaluating merits of potential solid breeders, other properties subject to different kind of investigations will enter into deciding in favor of an eventual solid breeder. Among them the stability of geometrical and mechanical properties under radiation is of paramount importance (17). Fig (21) and the following ones show pellets fabricated to test mechanical and thermal properties after irradiation.

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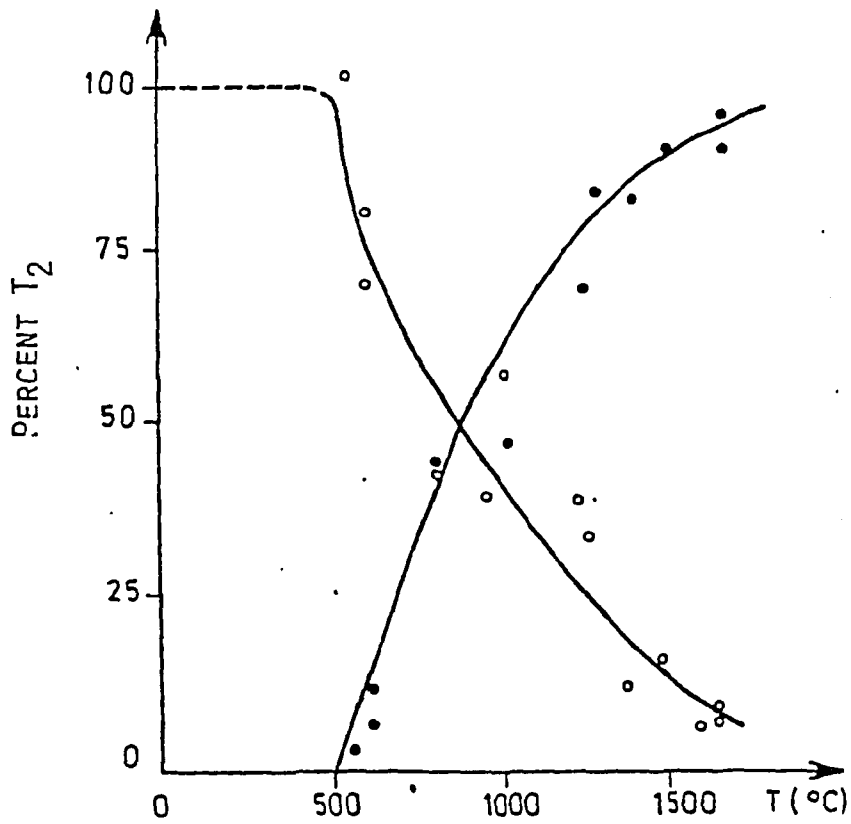
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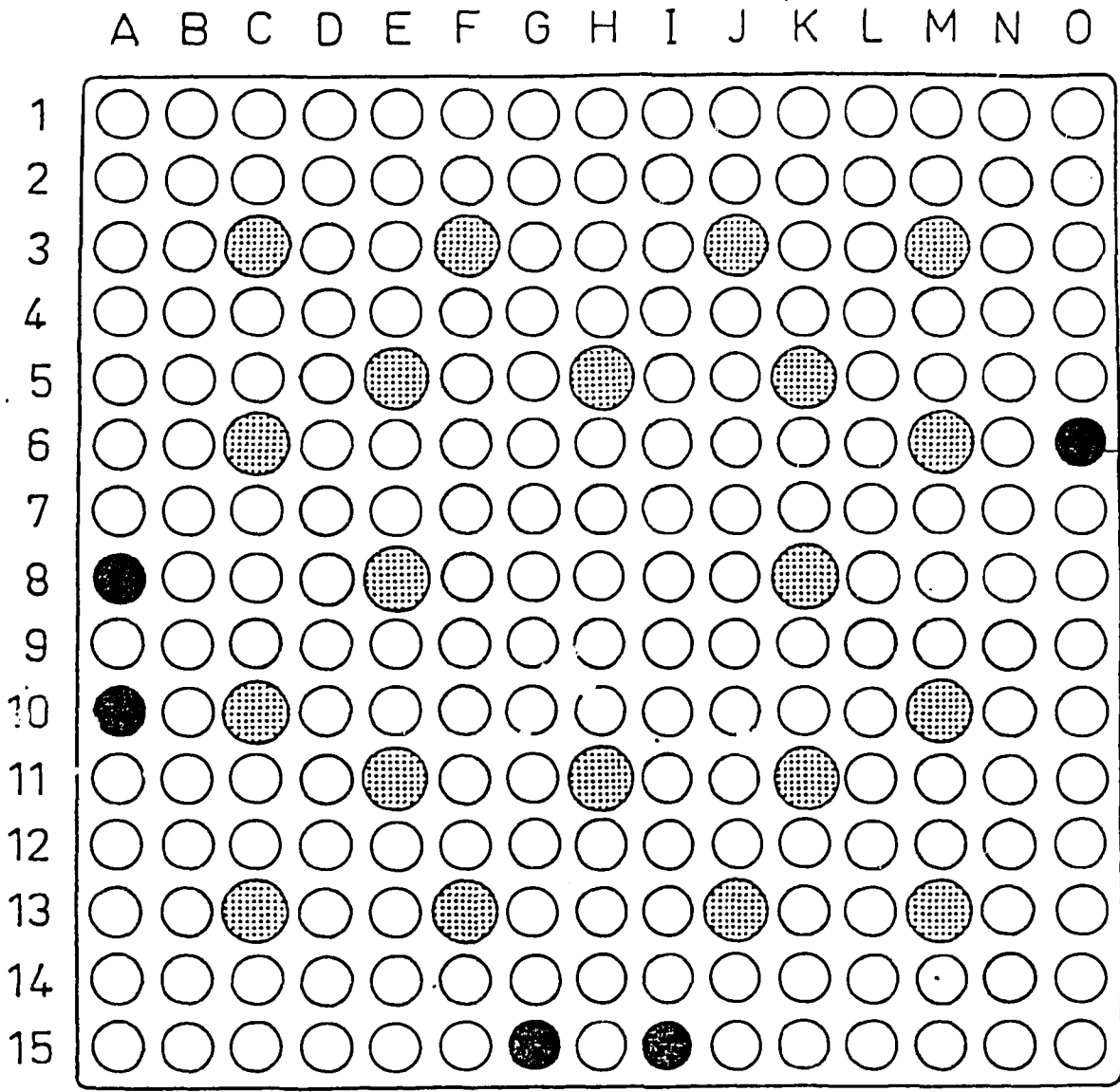
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FIGURE 1



PERCENT OF TOTAL  $T_2$  RETAINED IN  $UO_2$  (oo) AND IN ZIRCALOY (●●) AS A FUNCTION OF TEMPERATURE.

FIGURE 2



● guide    ○ Ordinary pin    ● Pin analysed

POSITION OF PINS OF THE BORSSLE FUEL ELEMENT.



FIGURE 3

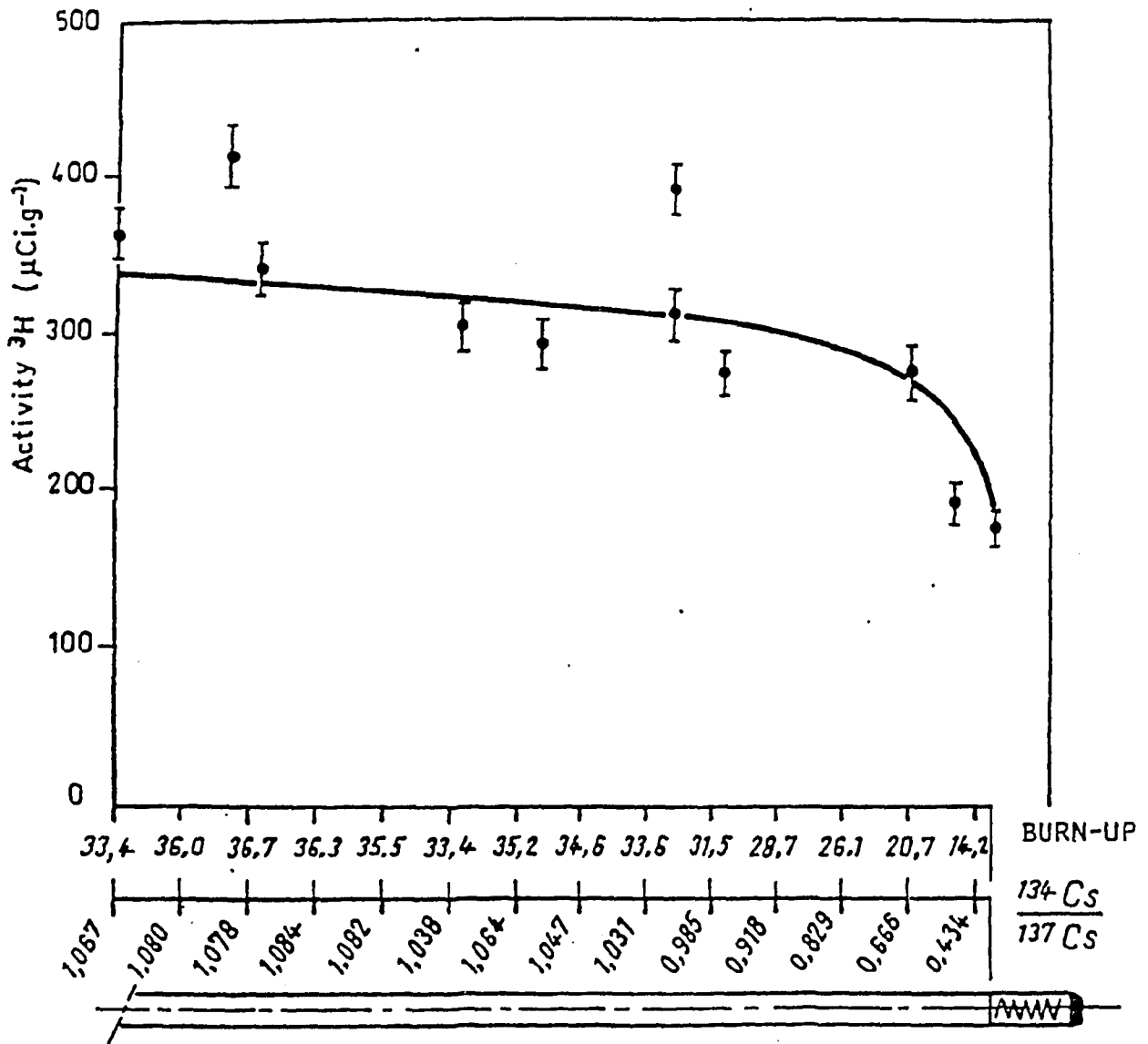


FIGURE 3 B

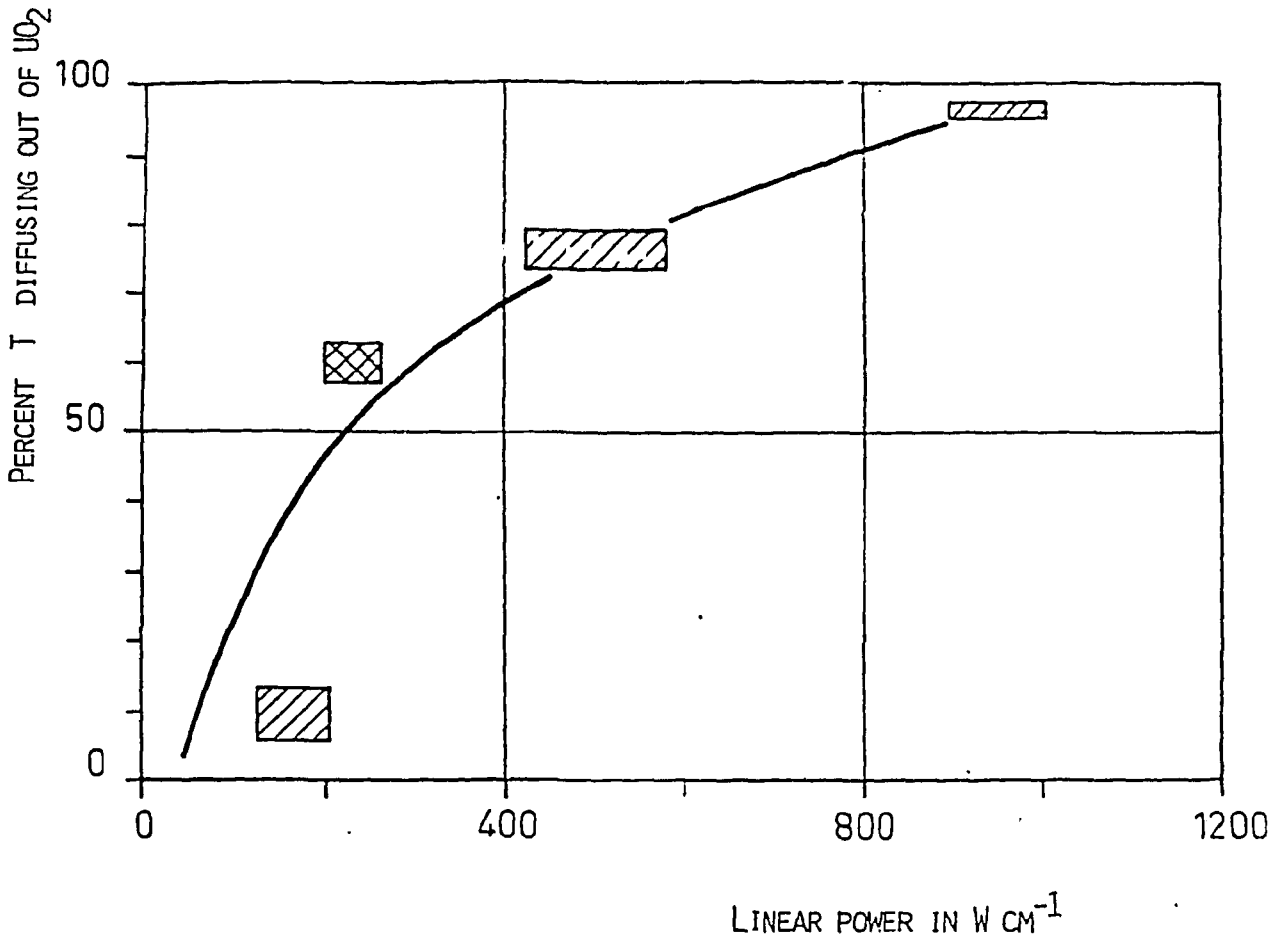


FIGURE 4 (CLICHÉ NON JOINT)

# "LILA" RIG

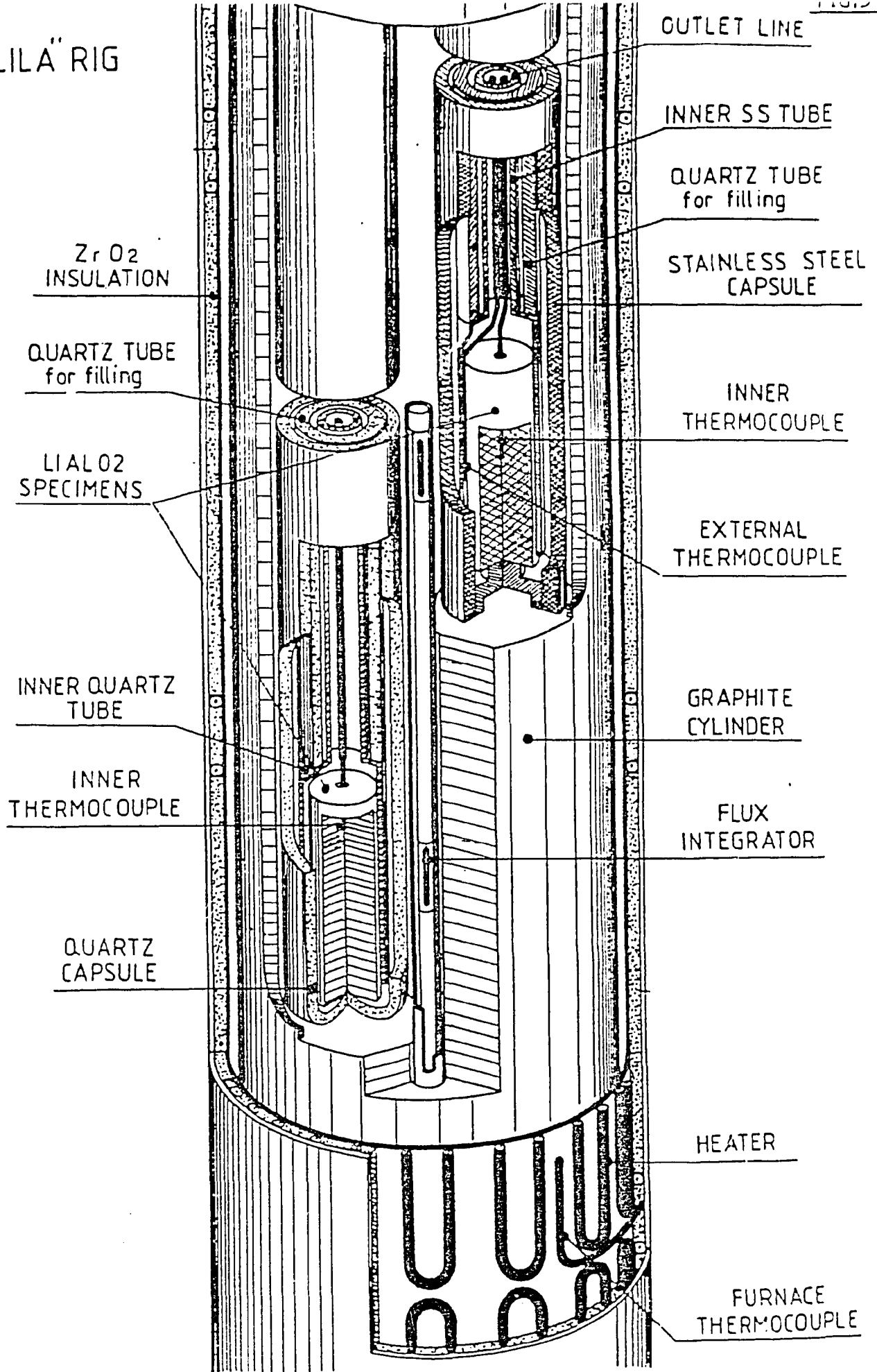
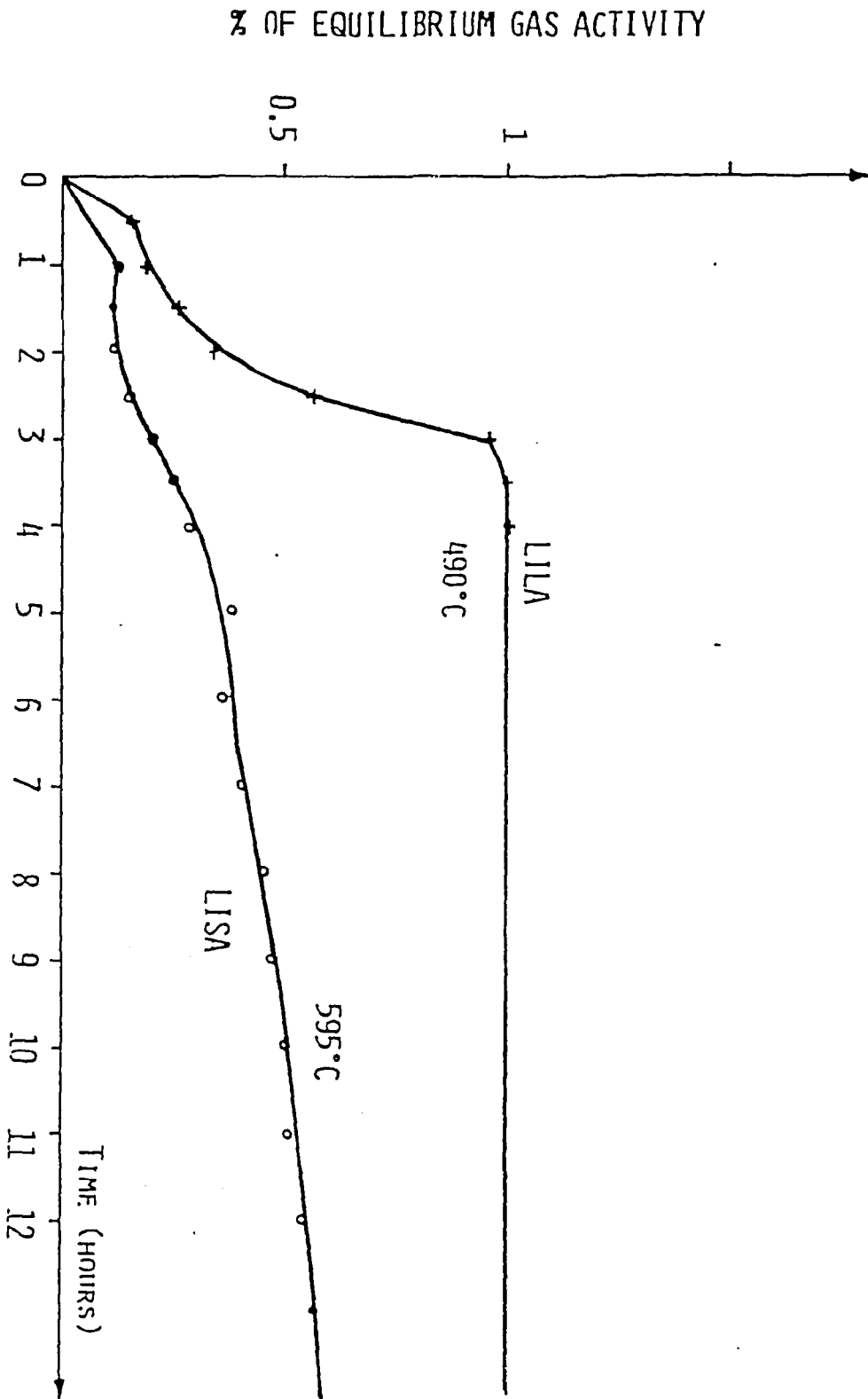


FIGURE 6



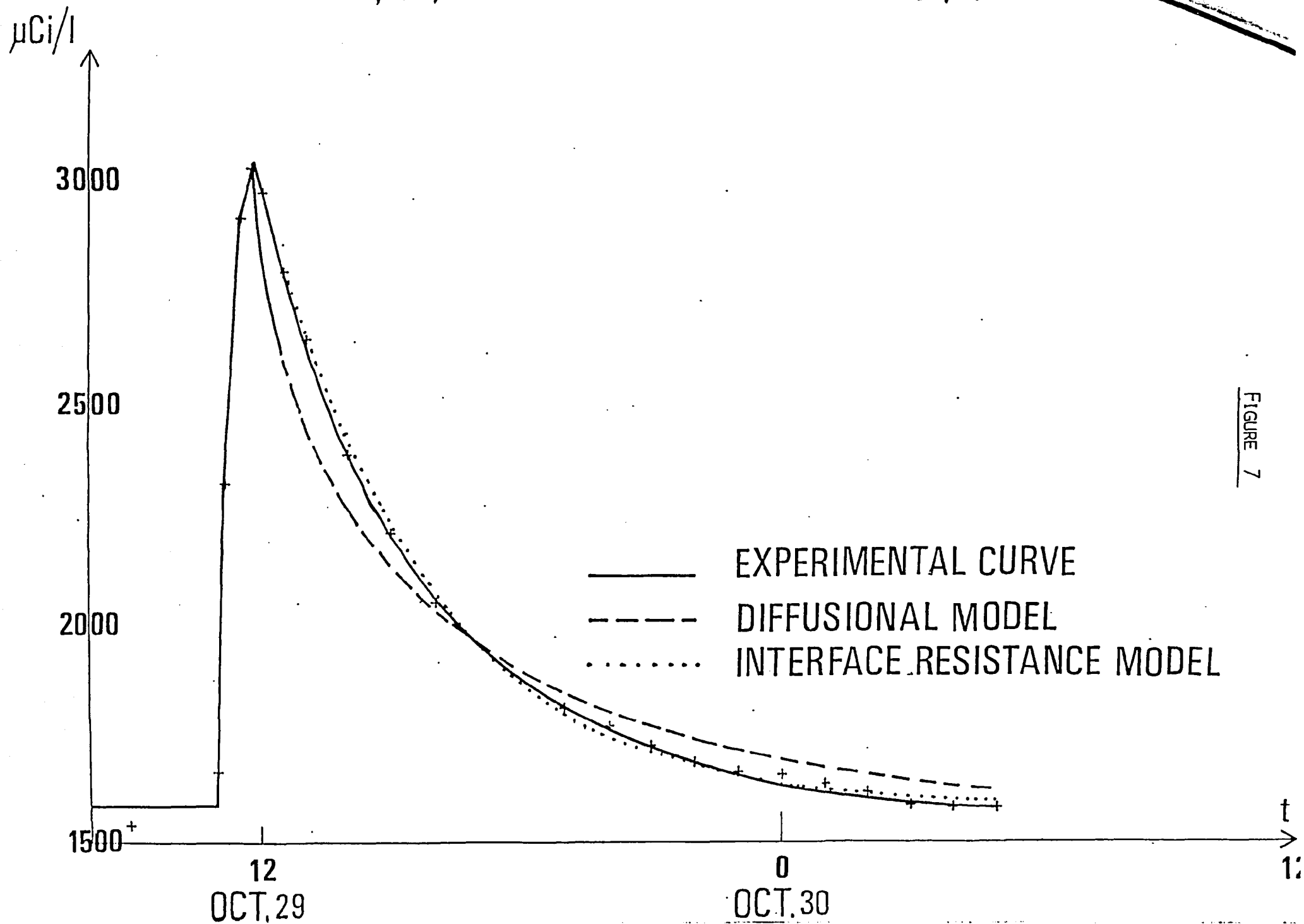


FIGURE 7

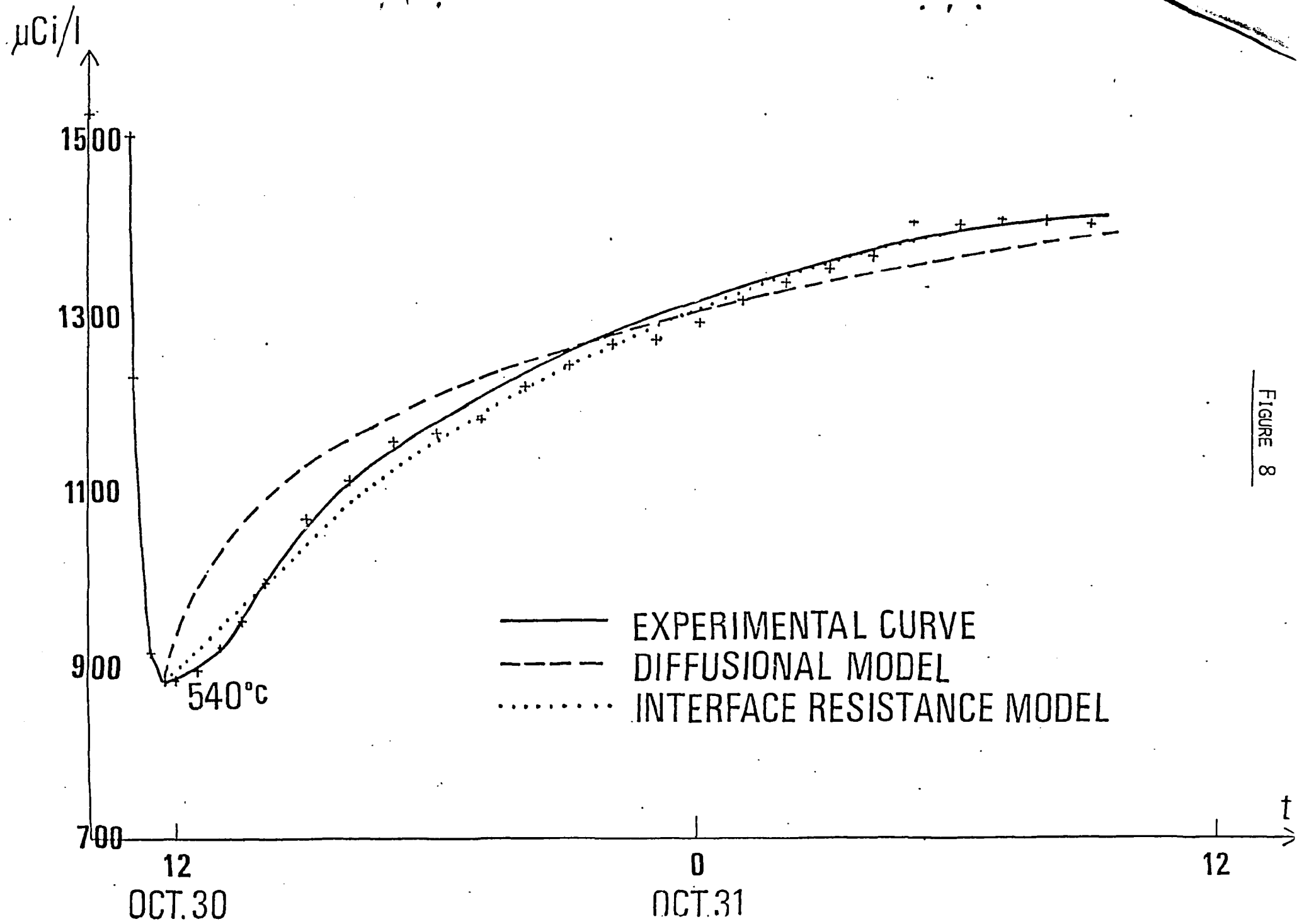
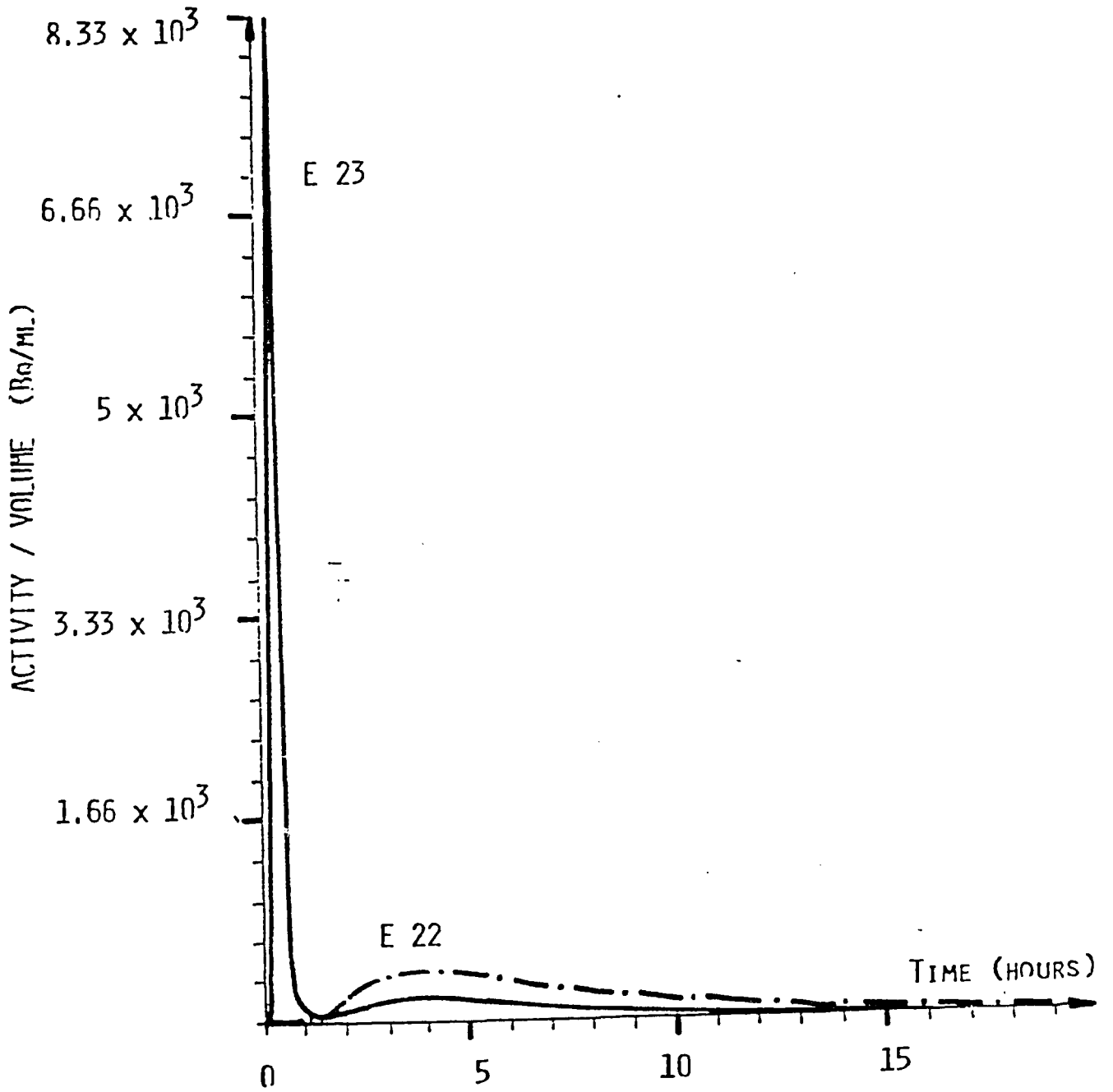
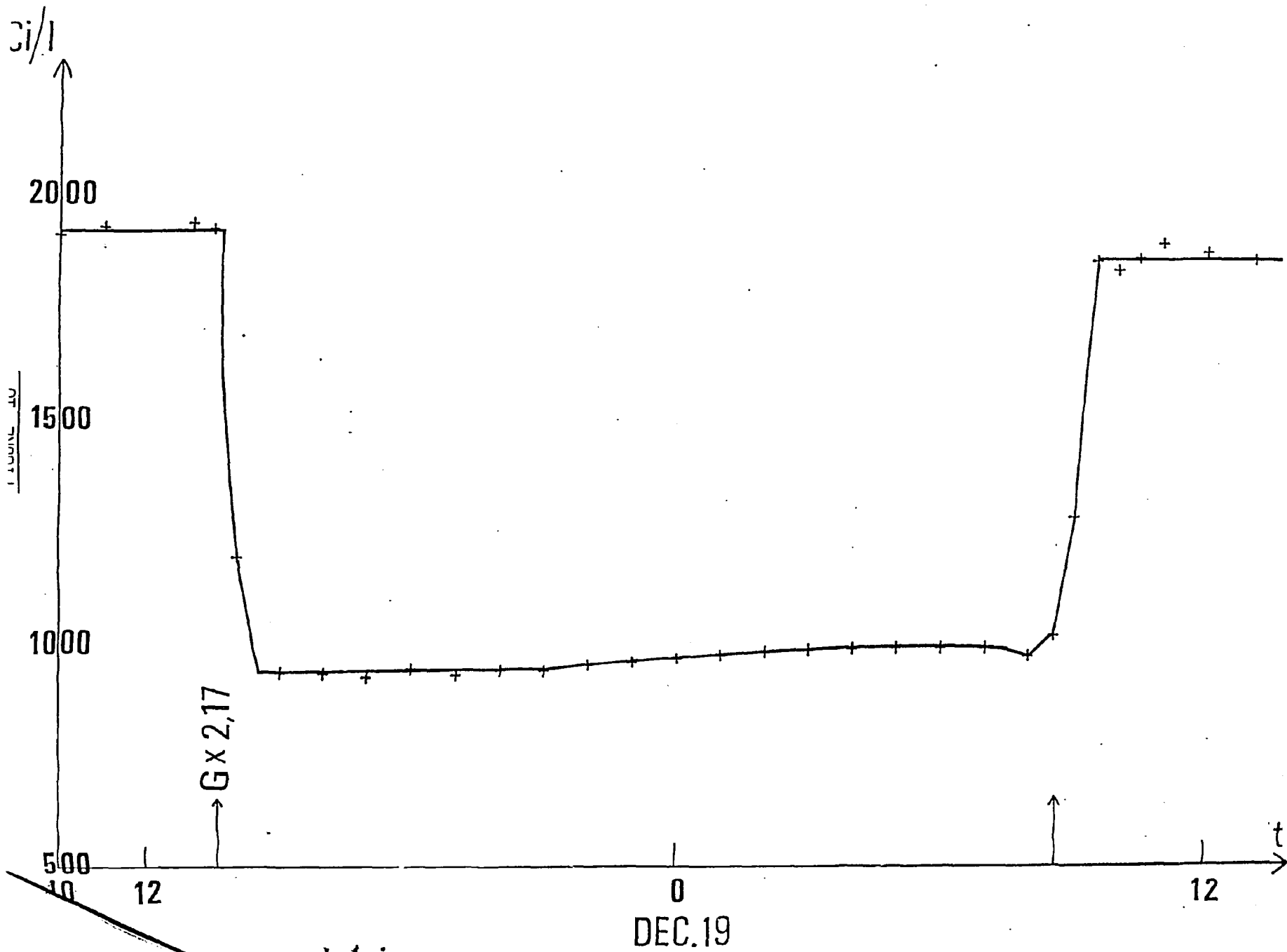


FIGURE 8

FIGURE 9







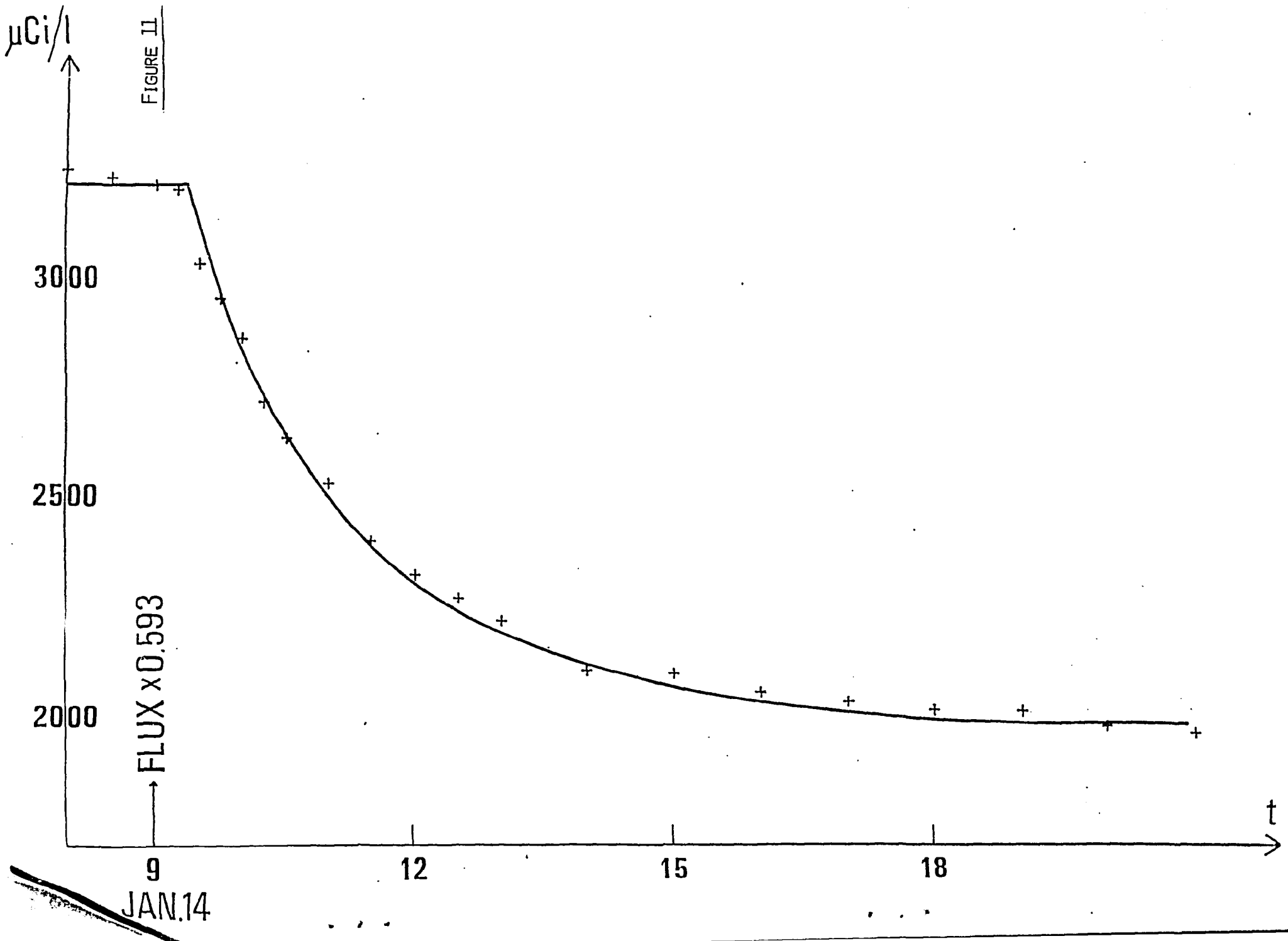


FIGURE 12

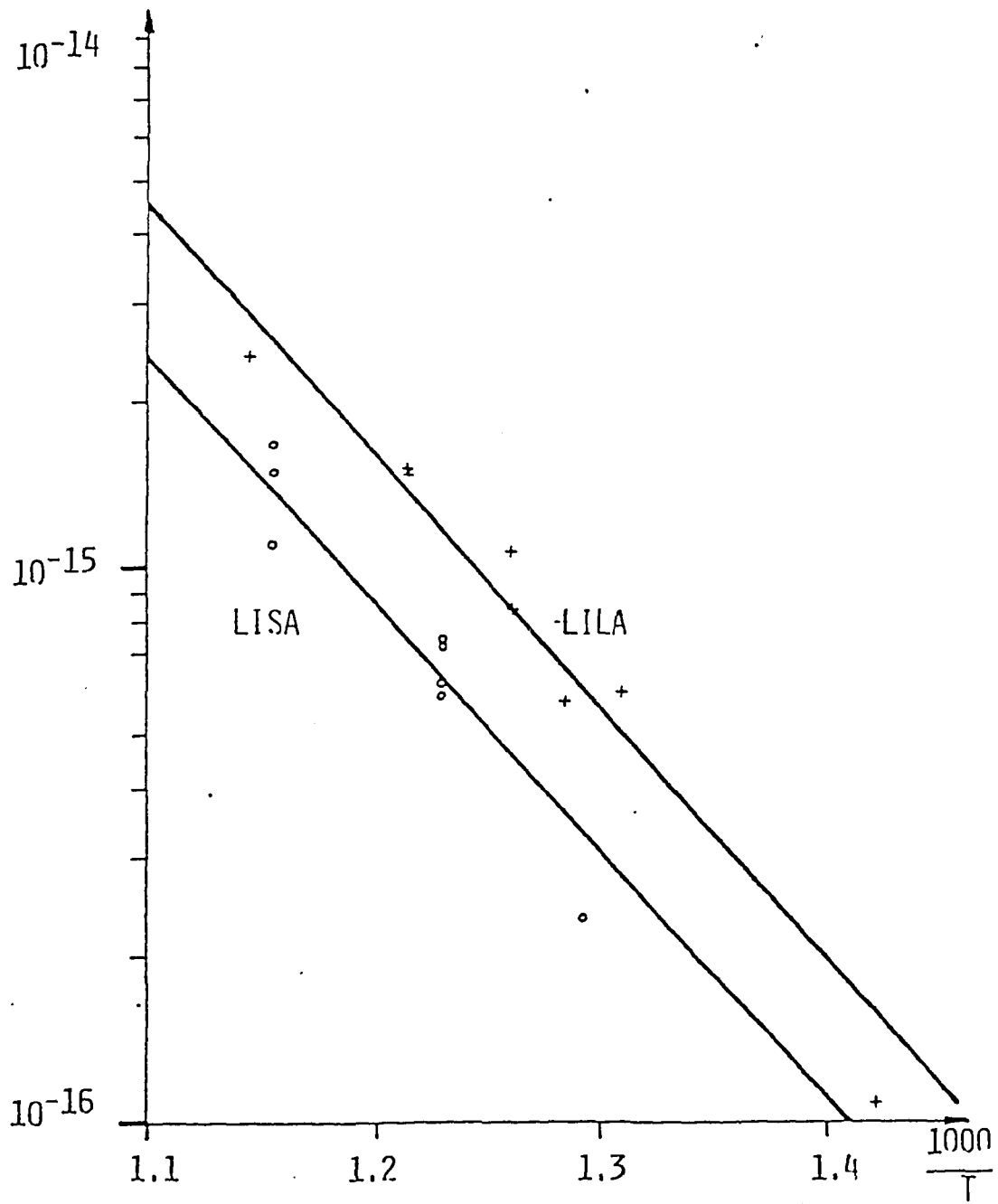


FIGURE 13

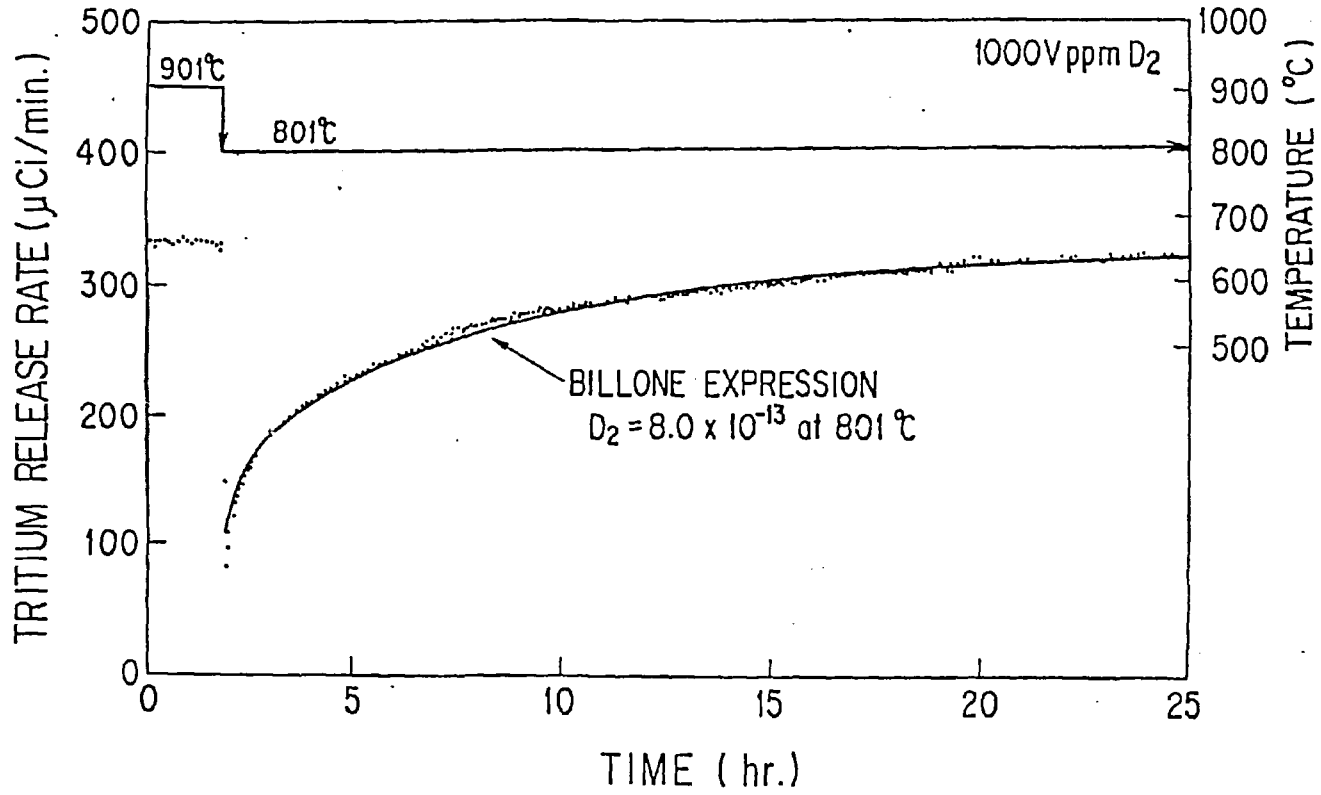


Fig. 6(a)

FIGURE 14

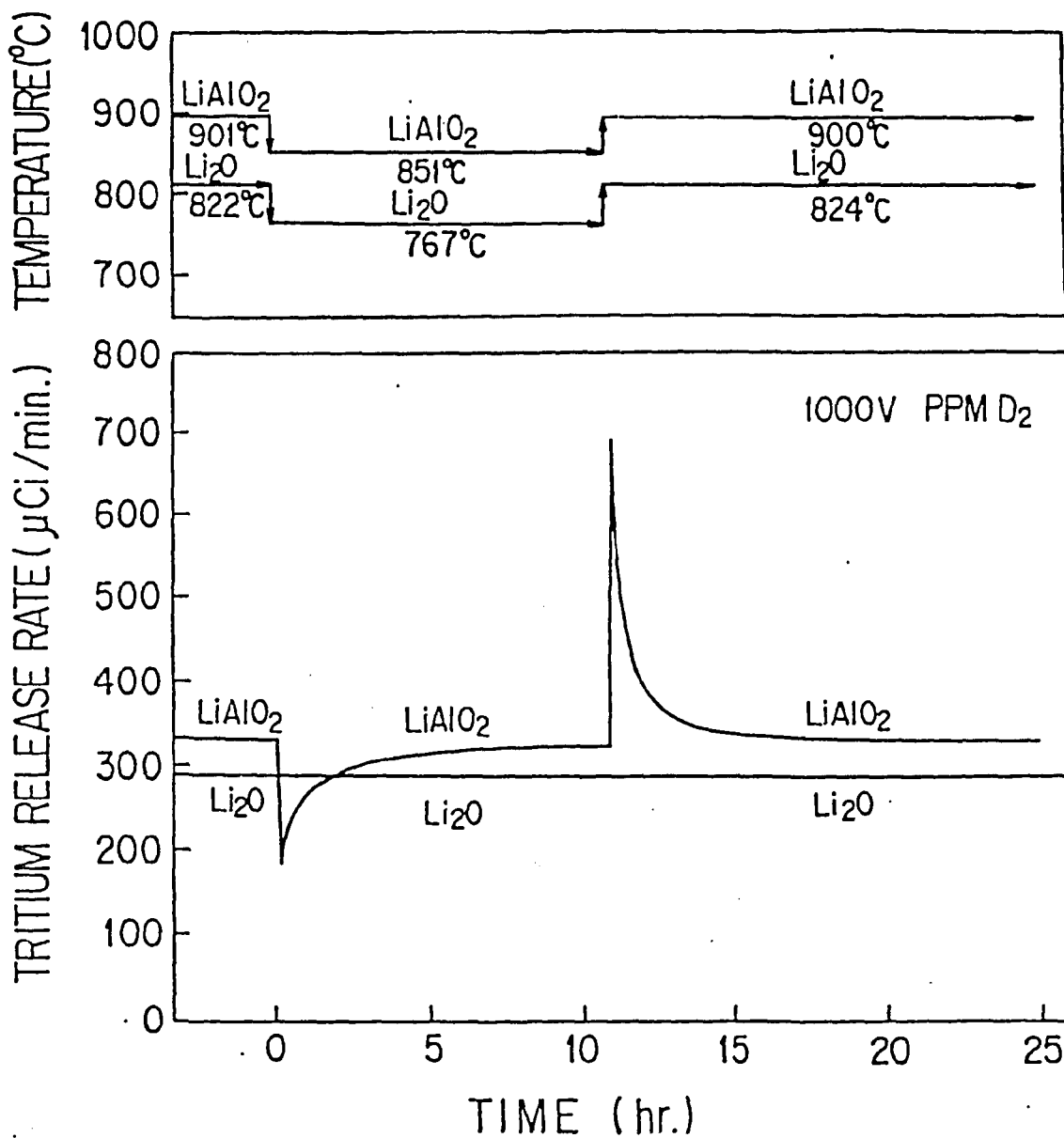


Fig.4(a)

FIGURE 15

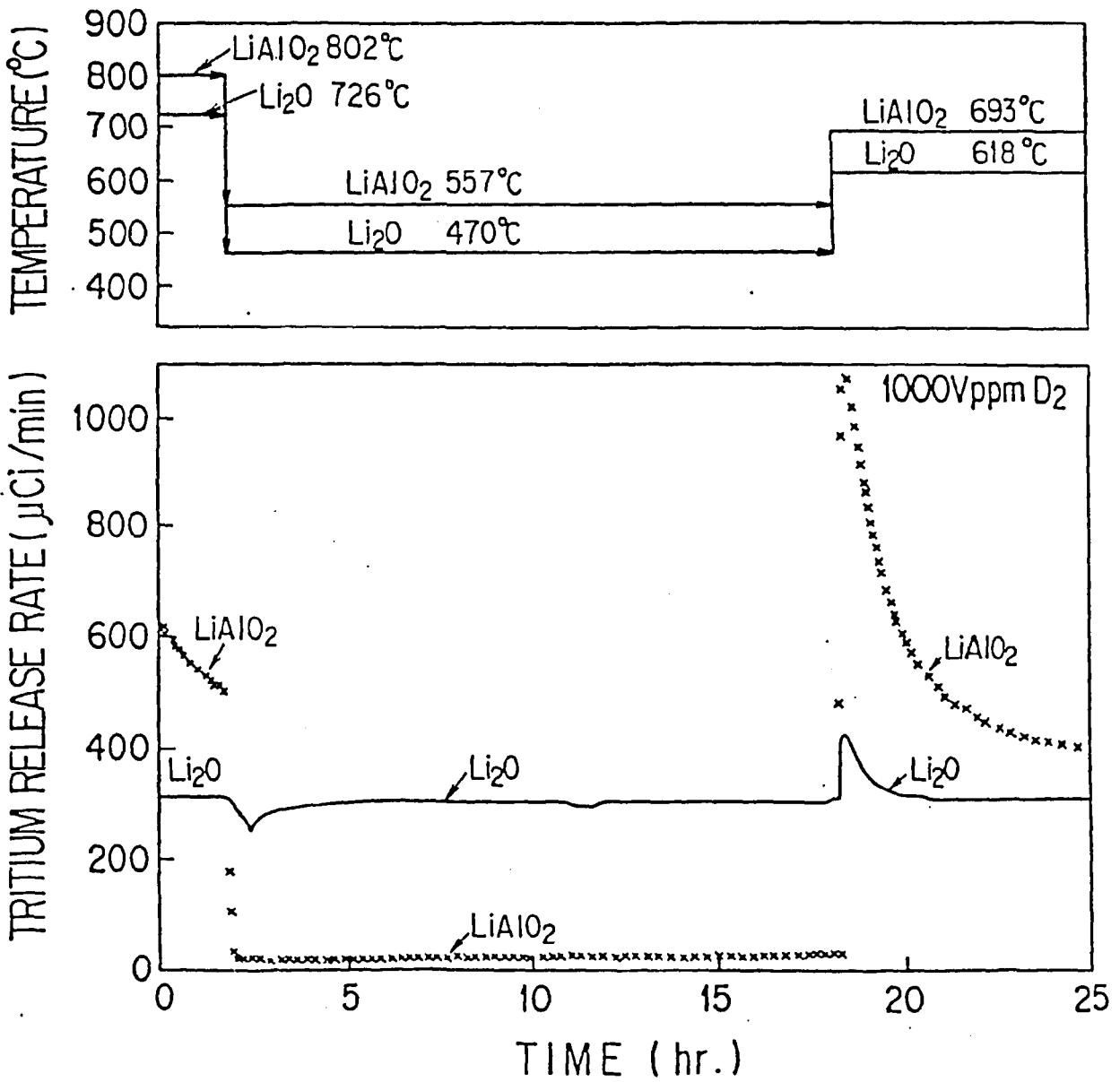
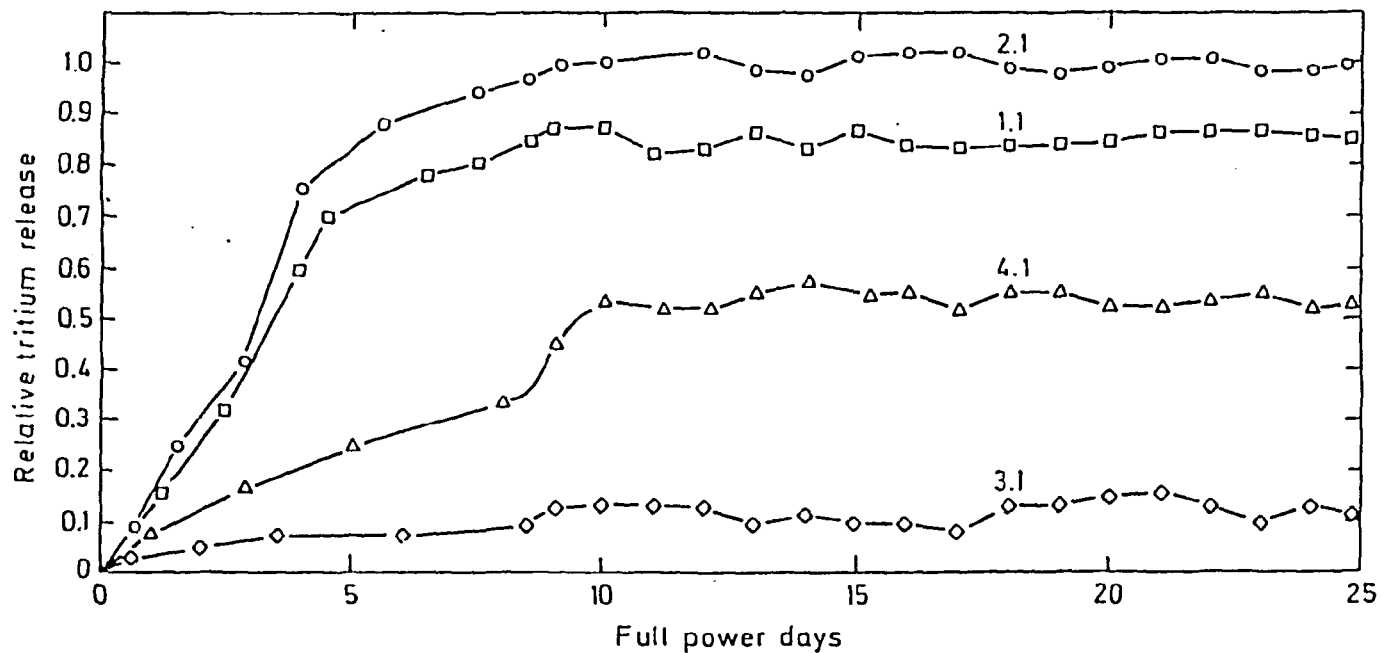


Fig. 4(b)

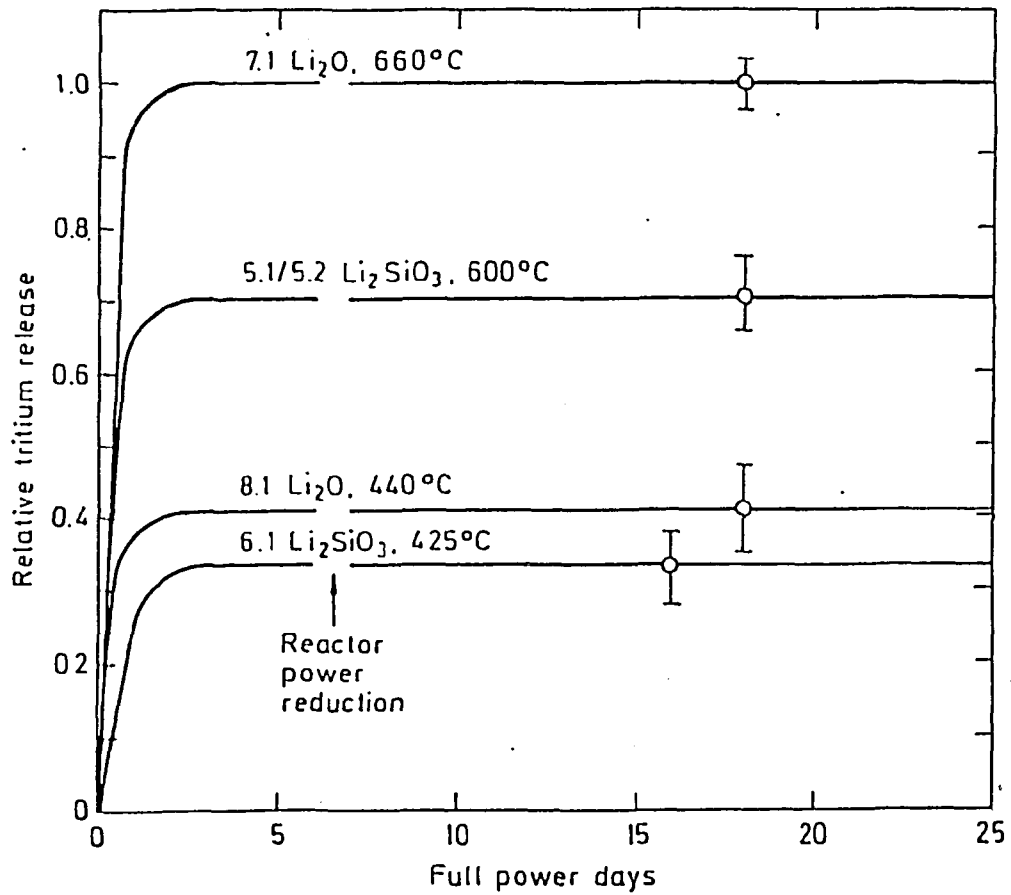
FIGURE 16



Relative tritium release of EXOTIC I

Curve	Sample	Average temperature	<sup>6</sup> Li content	% of tritium theoretical release rate
2-1	Li <sub>2</sub> SiO <sub>3</sub>	505°C	7.5 %	68
1-1	Li <sub>2</sub> SiO <sub>3</sub>	395°C	7.5 %	67
4-1	LiAlO <sub>2</sub>	460°C	7.5 %	96
3-1	LiAlO <sub>2</sub>	640°C	0.6 %	96

FIGURE 17



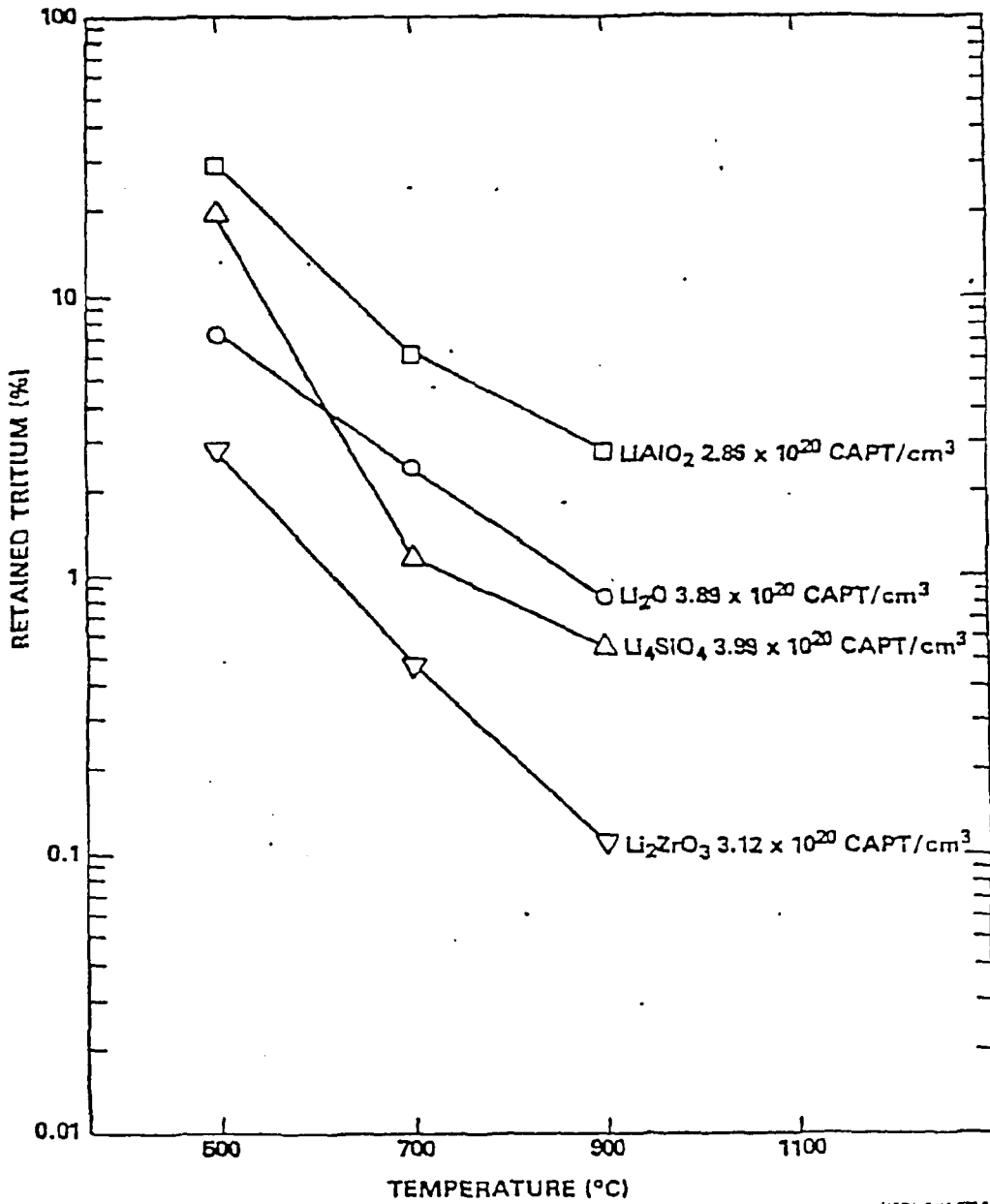
Curve	Tritium release : % of theoretical
7.1	108
5.1 + 5.2	44
8.1	51
6.1	32

All samples are 0.6% in  $^6\text{Li}$ .

Relative average tritium release of EXOTIC II



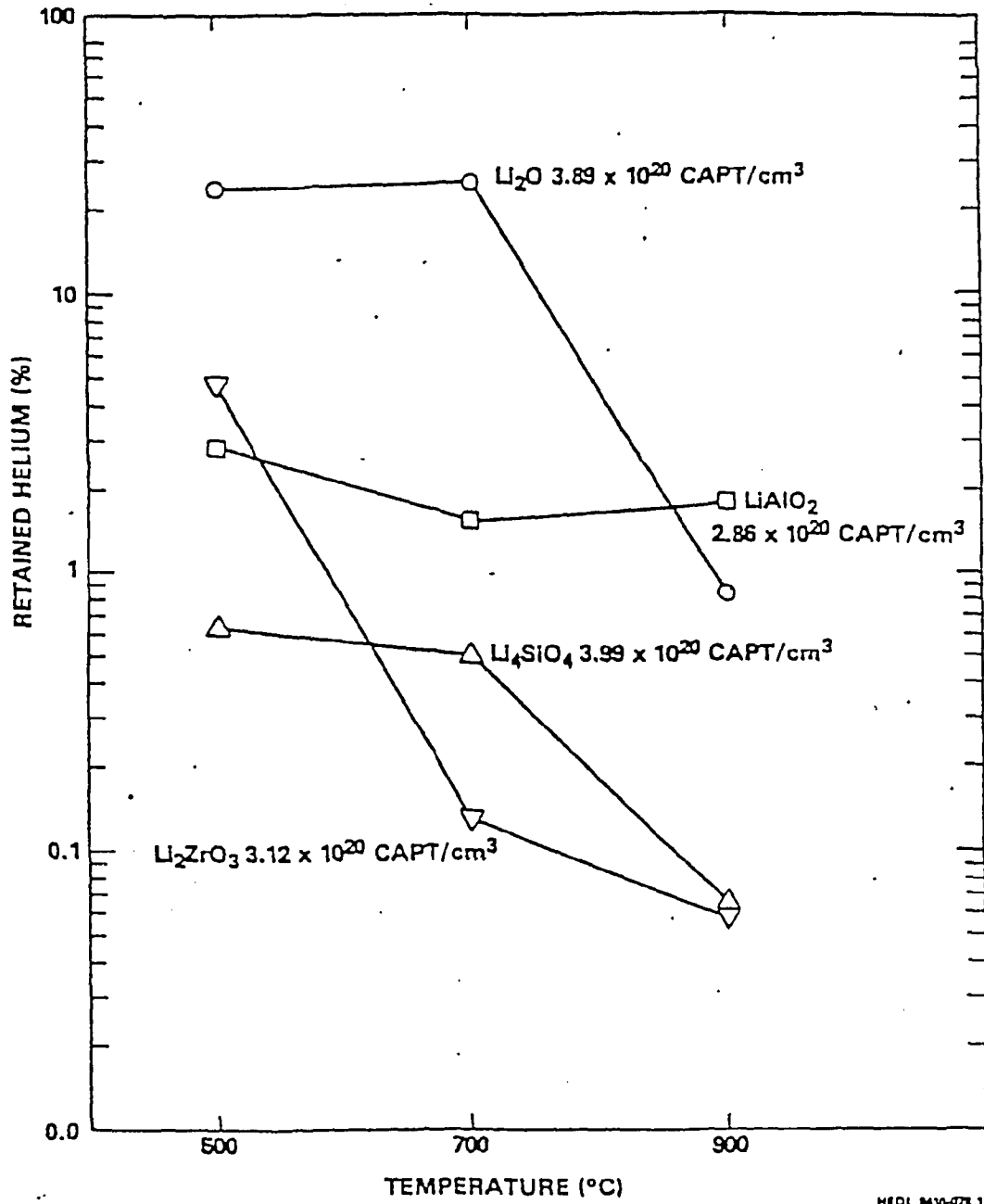
TRITIUM RETENTION IN  $\text{Li}_2\text{O}$ ,  
 $\text{LiAlO}_2$ ,  $\text{Li}_4\text{SiO}_4$  AND  $\text{Li}_2\text{ZrO}_3$



HEDL 8410-0762

Tritium Retention in  $\text{Li}_2\text{O}$ ,  $\text{LiAlO}_2$ ,  $\text{Li}_4\text{SiO}_4$  and  $\text{Li}_2\text{ZrO}_3$   
 After Irradiation.

FIGURE 19



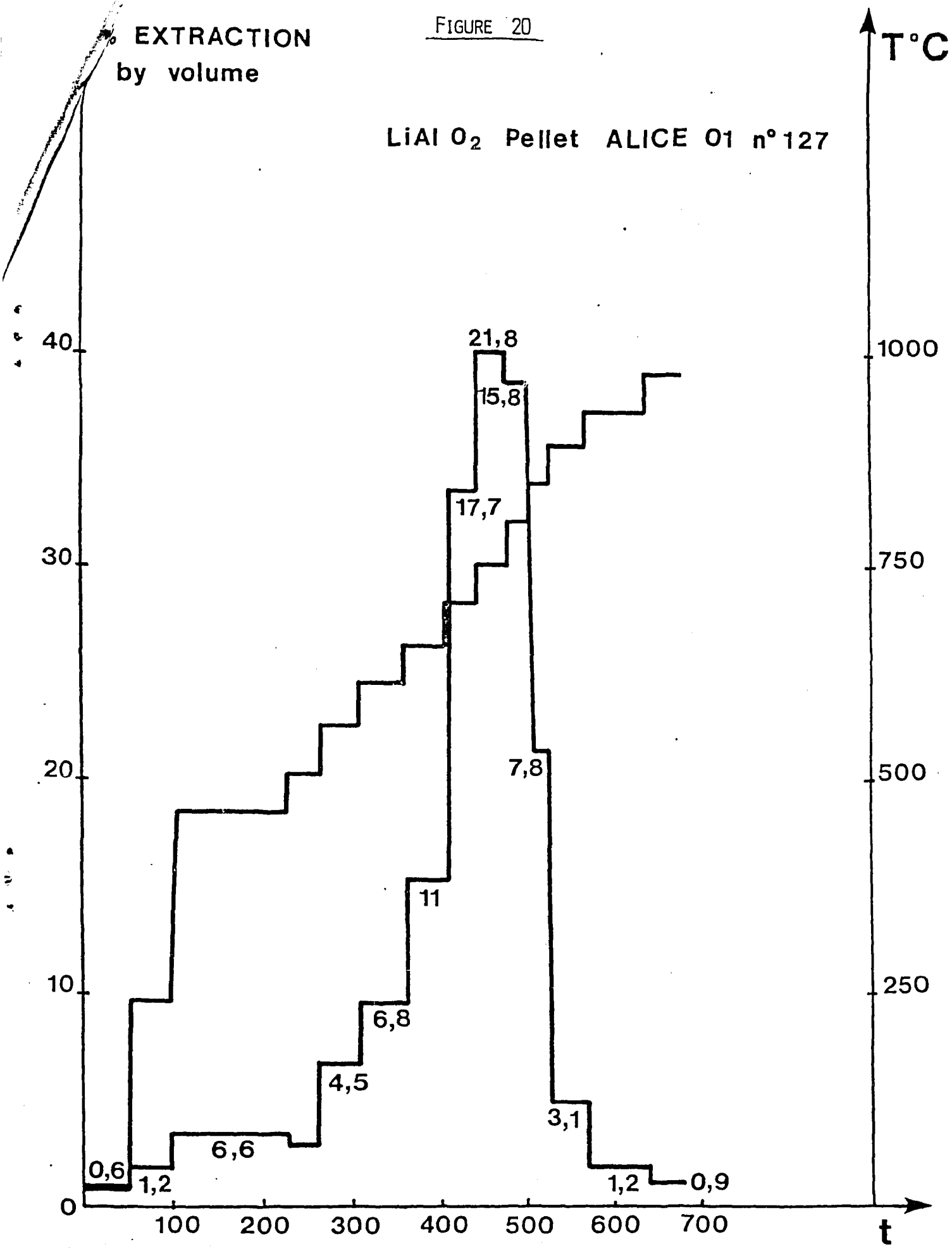
HEDL 8410-078.1

Helium Retention in Li<sub>2</sub>O, LiAlO<sub>2</sub>, Li<sub>4</sub>SiO<sub>4</sub> and Li<sub>2</sub>ZrO<sub>3</sub> After Irradiation.

FIGURE 20

EXTRACTION  
by volume

LiAlO<sub>2</sub> Pellet ALICE 01 n°127







FIGURE 21 (CLICHÉ NON JOINT)



T A B L E I

TRITIUM DISTRIBUTION BETWEEN URANIUM OXIDE AND ZIRCALLOY 4

HALF PIN	TOTAL TRITIUM ACTIVITY	TRITIUM IN THE FUEL	TRITIUM IN THE CLADDING	% OF TOTAL IN CLADDING
0 06 H	548	468	80	14
I 15 H	561	66	495	88
A 10 B	677	207	470	69
AVERAGE	595			
THEORETICAL	510			

NOTA : ALL ACTIVITIES ARE GIVEN IN ci FOR ONE TON OF FUEL,

H CORRESPONDS TO THE UPPER HALF OF A PIN , B TO THE LOWER ONE.

T A B L E II

AVERAGE ACTIVITIES OF TRITIUM EVOLVED IN THE GAS PHASE AND REMAINING IN SOLUTION WHEN DISSOLVING THE CLADDINGS (IN  $\mu$ CI PER GRAM)

HALF PIN	ACTIVITY IN THE GAS PHASE		ACTIVITY IN SOLUTION	% ACTIVITY IN THE GAS PHASE
	HT	CH <sub>3</sub> T		
0 6 H	79 $\pm$ 6	8 $\pm$ 0.7	219 $\pm$ 10	31 %
I 15 H	1896 $\pm$ 129	NOT MEASURABLE	9 $\pm$ 0.9	99.5 %
A 10 B	1799 $\pm$ 119	15 $\pm$ 0.9	8 $\pm$ 0.9	99.5 %

TABLE III

HULLS FROM PIN I 15 H	WEIGHT IN G.	TRITIUM ACTIVITY (*) IN THE GAS PHASE.		TRITIUM ACTIVITY (*) IN SOLUTION	TOTAL TRITIUM ACTIVITY(*)	% TRITIUM IN THE GAS FORM $\frac{3}{3}\text{HGAS}$
		HT	CH <sub>3</sub> T			
				(*) ALL ACTIVITIES EVALUA- TED AT THE END OF IRRADIA- TION.		$\frac{3}{3}\text{HGAS} + \frac{3}{3}\text{HDIS}$
E 12	2,20 ± 0,02	2071 ± 144	NON MEASURABLE	7,5 ± 0,5	2079 ± 145	99,6 %
E 13	2,16 ± 0,02	2078 ± 144	"	10,1 ± 0,6	2088 ± 145	99,5 %
E 14	2,12 ± 0,02	857 ± 60	"	5,2 ± 0,4	862 ± 61	99,4 %
E 15	2,17 ± 0,02	1921 ± 134	"	8,3 ± 0,5	1929 ± 135	99,6 %
E 16	2,18 ± 0,02	1228 ± 85	"	7,1 ± 0,6	1235 ± 86	99,4 %
E 17	2,14 ± 0,02	58 ± 4	"	0,8 ± 0,1	59 ± 4	98,7 %
E 18	2,20 ± 0,02	2063 ± 141	"	9,3 ± 0,9	2072 ± 142	99,5 %
E 19	2,19 ± 0,02	2114 ± 142	"	10,4 ± 0,9	2124 ± 143	99,5 %
E 20	2,22 ± 0,02	2121 ± 147	"	10,7 ± 0,9	2132 ± 148	99,5 %
E 21	2,28 ± 0,02	2066 ± 143	"	9,0 ± 0,9	2075 ± 144	99,5 %
E 22	2,82 ± 0,02	2160 ± 138	"	9,7 ± 0,8	2170 ± 139	99,5 %
E 23	2,24 ± 0,02	2182 ± 140	"	11,1 ± 0,9	2193 ± 141	99,5 %
AVE		1896 ± 129		9,0 ± 0,9	1095 ± 130	99,5 %

TABLE IV

HULLS FROM PIN A 10 B	WEIGHT IN G.	TRITIUM ACTIVITY (*) IN THE GAS PHASE.		TRITIUM ACTIVITY (*) IN SOLUTION	TOTAL TRITIUM ACTIVITY(*)	% TRITIUM IN THE GAS FORM $^3\text{H}_{\text{GAS}}$
		HT	$\text{CH}_3\text{T}$			
				(*) ALL ACTIVITIES EVALUATED AT THE END OF IRRADIATION.		$^3\text{H}_{\text{GAS}} + ^3\text{H}_{\text{DIS}}$
E 24	2.20 ± 0,02	1895 ± 126	1,6 ± 0,2	9.6 ± 1,1	1906 ± 127	99,5 %
E 25	2.17 ± 0,02	1758 ± 116	1,4 ± 0,2	8.3 ± 0,9	1768 ± 117	99,5 %
E 26	2.14 ± 0,02	1761 ± 118	1,2 ± 0,1	9.8 ± 1,1	1772 ± 119	99,4 %
E 27	2.12 ± 0,02	1605 ± 107	1,4 ± 0,2	7.1 ± 0,8	1613 ± 108	99,6 %
E 28	2.10 ± 0,02	2044 ± 138	1,4 ± 0,2	9.3 ± 1,0	2055 ± 139	99,6 %
E 29	2.01 ± 0,02	1749 ± 117	1,4 ± 0,2	8.2 ± 0,9	1759 ± 118	99,5 %
E 30	2.17 ± 0,02	2015 ± 133	1,4 ± 0,2	8.3 ± 0,9	2025 ± 134	99,6 %
E 31	2.10 ± 0,02	1869 ± 125	1,8 ± 0,2	7.9 ± 0,9	1879 ± 126	99,6 %
E 32	2.18 ± 0,02	1408 ± 92	1,2 ± 0,1	5.0 ± 0,5	1414 ± 93	99,6 %
E 33	2.16 ± 0,02	1940 ± 128	2,1 ± 0,2	7.6 ± 0,8	1950 ± 129	99,6 %
E 34	2.08 ± 0,02	1707 ± 113	1,6 ± 0,2	7.9 ± 0,8	1716 ± 114	99,5 %
E 35	2.13 ± 0,02	1829 ± 121	1,9 ± 0,2	7.1 ± 0,8	1838 ± 122	99,6 %
Ave		1799 ± 119	1,5 ± 0,2	8,0 ± 0,9	1808 ± 120	

TABLE V

HULLS FROM PIN CG H	WEIGHT IN G.	TRITIUM ACTIVITY (*) IN THE GAS PHASE.		TRITIUM ACTIVITY (*) IN SOLUTION	TOTAL TRITIUM ACTIVITY (*)	% TRITIUM IN THE GAS FORM $^3\text{HGAS}$
		HT	$\text{CH}_3\text{T}$			
E 1	$2,17 \pm 0,02$	$73 \pm 5$	$8,8 \pm 0,7$	$230 \pm 10$	$312 \pm 16$	26 %
E 2	$2,20 \pm 0,02$	$103 \pm 8$	$7,0 \pm 0,6$	$65 \pm 3$	$175 \pm 12$	63 %
E 3	$2,17 \pm 0,02$	$66 \pm 5$	$4,1 \pm 0,3$	$343 \pm 15$	$413 \pm 21$	17 %
E 4	$2,22 \pm 0,02$	$84 \pm 6$	$13 \pm 1$	$268 \pm 12$	$365 \pm 19$	26 %
E 5	$2,16 \pm 0,02$	$87 \pm 6$	$14 \pm 1$	$243 \pm 12$	$344 \pm 19$	29 %
E 6	$2,23 \pm 0,02$	$86 \pm 6$	$7,0 \pm 0,6$	$100 \pm 5$	$193 \pm 12$	48 %
E 7	$2,19 \pm 0,02$	$70 \pm 5$	$8,6 \pm 0,7$	$314 \pm 15$	$393 \pm 21$	20 %
E 8	$2,09 \pm 0,02$	$75 \pm 5$	$7,1 \pm 0,6$	$194 \pm 9$	$276 \pm 15$	30 %
E 9	$2,24 \pm 0,02$	$86 \pm 6$	$4,9 \pm 0,4$	$187 \pm 10$	$278 \pm 17$	33 %
E 10	$2,19 \pm 0,02$	$70 \pm 5$	$6,3 \pm 0,5$	$247 \pm 12$	$323 \pm 18$	24 %
E 11	$2,20 \pm 0,02$	$65 \pm 4$	$7,8 \pm 0,6$	$218 \pm 10$	$291 \pm 15$	25 %
AVE		$79 \pm 6$	$8,0 \pm 0,7$	$219 \pm 10$	$306 \pm 17$	31 %

(\*) ALL ACTIVITIES EVALUATED AT THE END OF IRRADIATION.

$^3\text{HGAS} + ^3\text{HDIS}$



TABLE VI

Diffusion inventories, purge gas He+0.1% H<sub>2</sub>

Run	Temp. (°C)	P6, Orthosilicate, p=4.76 mC/h				P2, Aluminate, p=4.80 mC/l		
		I (mC)	$\tau$ (h)	D (cm <sup>2</sup> /s)		I (mC)	$\tau$ (h)	D (cm <sup>2</sup> /s) r=.19 $\mu$
r=13 $\mu$	r=.4 cm							
B4	500	22.3	4.7	$.7 \cdot 10^{-11}$	$.7 \cdot 10^{-6}$	41.9	8.7	$.8 \cdot 10^{-15}$
C2-7	550	8.6	1.8	$1.7 \cdot 10^{-11}$	$1.7 \cdot 10^{-6}$	11.3	2.4	$2.8 \cdot 10^{-15}$
	600	5.7	1.2	$2.6 \cdot 10^{-11}$	$2.6 \cdot 10^{-6}$	3.6	.8	$8.3 \cdot 10^{-15}$
	630	5.1	1.1	$2.8 \cdot 10^{-11}$	$2.8 \cdot 10^{-6}$	2.7	.6	$11 \cdot 10^{-15}$

p production rate, estimated from steady - state release

I inventory

 $\tau=I/p$  residence time $D=r^2/15\tau$  diffusion coefficient, r characteristic radius for diffusion