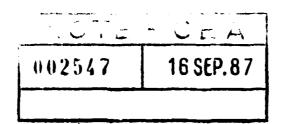
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### COMMISSARIAT A L'ENERGIE ATOMIQUE

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## Institut de Recherche Technologique et de Developpement Industriel

# DIVISION D'ETUDES DE SEPARATION ISOTOPIQUE ET DE CHIMIE PHYSIQUE



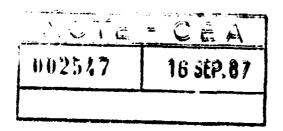
### **FUSION TECHNOLOGY PROGRAM**

TRITIUM TECHNOLOGY

PROGRESS REPORT

# TASK ACTION $T_2$ A HYDROGEN EXTRACTION FROM A GAZ MIXTURE

### M. CARON-CHARLES



### TASK ACTION T2A

## HYDROGEN EXTRACTION FROM A GAZ MIXTURE ABSTRACT

Within the framework of the european fusion program, we are dealing with the tritium technology aspect.

Burned gases exhausted from a plasma chamber contain impurities linked with hydrogen atoms like  $CH_4$ ,  $NH_3$ ,  $H_2O$ ... After being separated from the main hydrogen stream, theese impurities will be decomposed to recover their tritium content.

Hydrogen, free or under a combined form within a  $\rm H_2$ ,  $\rm N_2$ ,  $\rm NH_3$ ,  $\rm CH_4$ ,  $\rm O_2$  gaseous mixture, can be purified by chemical reaction with uranium metal. The resulting reaction consists in absorbing the impurities without holding back  $\rm H_2$ . Working conditions have been defined according to two main goals: the formation of stable solid products, especially under hydrogenated atmosphere, and the optimization of the material quantities to be used.

Thermodynamic and kinetic considerations have shown that the 873 K - 1173 K temperature range should be suitable for such reactions, assuming however that  $\mathrm{CH_4}$  or  $\mathrm{NH_3}$  are not diluted in a great excess of  $\mathrm{H_2}$ . Experiments performed at 973 K have produced hydrogen according to the predicted reaction rates. But they have also pointed out the importance of interferences that might occur in the uranium-gas system, on the gases conversion rates.

### HYDROGEN EXTRACTION FROM A GAZ MIXTURE

### M. CARON-CHARLES

### 1.- INTRODUCTION.

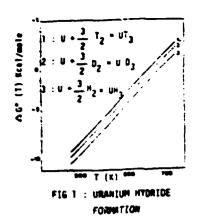
Within the framework of the european project of thermonuclear fusion, we are dealing with the tritium technology aspect.

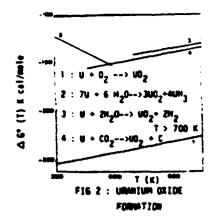
Burned gases exhausted from a plasma chamber contain impurities linked with hydrogen atoms like  $\mathrm{CH_4}$ ,  $\mathrm{NH_3}$ ,  $\mathrm{H_2O}$ ... After being separated from the main hydrogen stream theese impurities will be decomposed to recover their tritium content. The aim of our work is to study the reduction of hydrogenated molecules by chemical reaction with an appropriate metal. It consists in absorbing impurities without holding back hydrogen.

The chemical properties of uranium let us suppose that it could be applied to the cracking of hydrogenated molecules and to purified hydrogen production. Working conditions have to be defined according to two main goals: the formation of solid products, especially under hydrogenated atmosphere, and the optimization of the material quantities to be used.

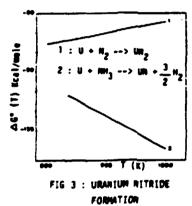
### 2.- THERMODYNAMIC STUDY OF THE CHEMICAL REACTIONS.

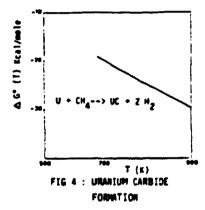
For T > 423 K, The hybride formed is UH<sub>3</sub> ( $\beta$ ) and respectively UD<sub>3</sub> ( $\beta$ ) and UT<sub>3</sub> ( $\beta$ ), with deuterium and tritium [1]. The equilibrium reaction between uranium and hydrogen, implies that, at T=700K if hydrogen pressure (respectively deuterium and tritium pressure) is lower than 1.31  $10^5$  Pa (respectively 1.99  $10^5$ Pa and 2.66  $10^5$  Pa) no uranium hydride will be formed (Fig.1).





The two stable forms of uranium oxide are  ${\rm UO_2}$  and  ${\rm U_3~O_8~I2!}$ . The first step of uranium oxidation being  ${\rm UO_2}$  formation, we only take this reaction into account in the thermodynamic study. All the oxidation reactions are irreversible ( Fig. 2 \cdot).





The three stable forms of uranium nitride being UN,  $U_2$   $N_3$  and  $UN_2$ , the first step of the reaction is the formation of  $UN_2$  for T < 1000 K and of UN for T > 1000 K i3, 41. Only UN formation was considered since no thermodynamic values were found for  $UN_2$ . UN form is the most stable one. At 1473 K, at pressure lower than  $10^{-4}$  Pa, it could be decomposed into uranium and gaseous nitrogen, when 0.25 Pa is sufficient to decompose  $U_2$   $N_3$  or  $UN_2$  as  $U_2$   $N_3$  --->  $2UN_3$  +  $\frac{1}{2}$   $N_2$  | 3,41. The results gathered in figure 3, confirm that nitride formation is irreversible.

Like oxide and nitride productions, the reaction between uranium and methane is irreversible (Fig 4), the first step being the formation of the monocarbide UC  $^{\dagger}$ 5.

We have been interested by the chemical reactions between the reactions products: oxides (  $U0_2$ ,  $U_3$   $0_8$  ), nitrides (  $UN_2$ ,  $U_2$   $N_3$  ) and uranium carbides ( UC,  $UC_2$  ) and a fresh gaseous mixture of  $H_2$ ,  $O_2$ ,  $NH_3$ ,  $CH_4$ ,  $N_2$ . A bibliographic investigation suggests that hydrogen reduction as well as oxidation of the solid products are the two important effects [7].

All the products formed with pure uranium may be oxidized, giving off impurities such as  ${\rm CO_2}$ ,  ${\rm O_2or~N_2}$ , even with oxygen traces, as soon as Kp is lower than  ${\rm K_p}$  eq. (Table 1 ).

TABLE 1 : ORIGINATION OF THE SOLID

I Rection	6" (600K)   t cal/mp1	in r
3 W02 + 02	- 64.73	\$4
) w • 4 0 <sub>2</sub>	- 527	439
3 02 N3 0 05 9 N2	- 1070	892
3 0 C + 7 0 <sub>2</sub> > 0 <sub>3</sub> 0 <sub>0</sub> + 3 C0 <sub>2</sub>	-611	676

The hydrogenated molecules being cracked, the reactions products will be in contact with great quantity of free hydrogen, giving rise to new chemical reactions (Fig 5).

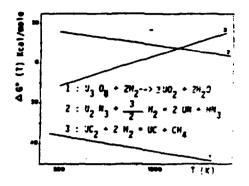


FIG 5 : REDUCTION OF THE SOLID PRODUCTS BY H<sub>2</sub>

Hydrogen completely reduces  $\rm U_3$   $\rm O_8$  from the low temperatures giving off  $\rm H_2O$ . The equilibrium reactions of  $\rm U_2$   $\rm N_3$  and  $\rm UC_2$  reductions prove that temperature and pressure conditions have to be judiciously choosen, in order to avoid any ammonia either methane releasing, this occurring respectively at the highest temperature and at the lowest one. For example, with an hydrogen pressure of  $10^5$  Pa, the releasing of ammonia can reach 1.29  $10^4$ Pa at 1300 K and 3.11  $10^4$  Pa pressure of CH<sub>4</sub> at 900K, at the equilibrium.

The reactions between the solid compounds imply that oxygen, nitrogen and carbon have to be dissolved in the same uranium lattice, to give U (C, O, N). This occurs at T > 1473 K and  $P < 7 \cdot 10^{-3}$  Pa, and no significant gaseous releasing will be observed |6|.

These thermodynamic considerations suggest us that uranium could be used for the extraction of free hydrogen from the gaseous mixture  $H_2$ ,  $N_2$   $NH_3$ ,  $CH_4$ ,  $O_2$ , but under appropriate operating conditions of temperature and pressure.

At temperature higher than 700 K and hydrogen pressure lower than 1.31  $10^5$  Pa, hydrogen will not react chemically. On the contrary, the remaining gases will form solid uranium products. They could however be oxidized giving off N<sub>2</sub> or CO<sub>2</sub>.A solution might be to favour oxidation of pure uranium in order to eliminate any oxygen trace. They could also be reduced by hydrogen giving off H<sub>2</sub> O, CH<sub>4</sub> or NH<sub>3</sub>, this occurring preferably at T < 900 K and T > 1200 K for carbides and nitrides respectively, and as soon as K<sub>p</sub> is lower than the equilibrium constant K<sub>p</sub>.

The practical use of uranium under these conditions depends however on the kinetics of the reactions: the initial solid attack, representative of surface phenomena, the kinetics at high conversion rate of the solid, that will give the usable mass of solid, and the likely interferences that should be taken into account to obtain a good representation of the system uranium-gases mixture. The two first points were previously described in litterature [2,5,8,9]. In order to define the resistance of uranium to gases such as oxygen, ammonia, nitrogen or methane, the authors carried out their investigations at constant pressure. Solid attacks are generally linear and produce porous layers that can also undergo desquamation. The results agree with the possible formation of a compact film of product, laying on the uranium surface (2,3,5). Diffusion of gas through this compact layer could be the limiting step of each chemical reaction and will be related to the evolution of composition and compacity of the reaction products with temperature.

### 3.- EVOLUTION OF THE NATURE OF THE PRODUCTS WITH TEMPERATURE.

At 573 < T < 973 K, the formation of  $\rm U_3~O_8$  favours uranium oxidation by fissuring the product layer, so that the compact film  $\rm UO_2$  is no more protective |2| - At T > 1073 K,  $\rm U_3O_8$  sintered and oxidation rates slow down |7|.

At T < 1023 K, UN<sub>2</sub> is the main product in the compact film and in the out-layer solid. From 1023 K to 1173 K, there would be a film of UN and an out-layer of  $U_2$  N<sub>3</sub> -UN<sub>2</sub> [3,4], and the solid is as more pulverulent as the temperature is growing up [8,9].

At T < 973 K, uranium methane reaction mainly provides UC, while at T > 1173 K, pure  $UC_2$  may be formed, the reaction rate increasing with temperature |5|.

These thermodynamic and kinetic results let us suggest the existence of a temperature range 873 K < T < 1173 K, where impurities consumption as well as products stability under various conditions should be the most efficient.

### 4.- PRELIMINARY EXPERIMENTS.

In order to check and extend the experimental results established in litterature and more precisely to define the interferences that may occur in uranium-gases mixture system, we have built an apparatus (Fig 6).

Gases mixture, prepared in a first loop, is flowing in the reactor loop, which volume is 2.5 liters, at rates set at about 5 1/mn. Thermocouples (type K) measure solid and gas temperature respectively at  $T_1$  and  $T_{2-->5}$ , capacity transmitters of pressure provide gas pressure at  $P_1$  and  $\Delta P_1$ , and a gas chromatograph define gas composition as function of time. We utilize small cylinders, cut in impoverished uranium of 99.5 % purity, that we first clean with pure nitric acid and then polish with abrasive paper.

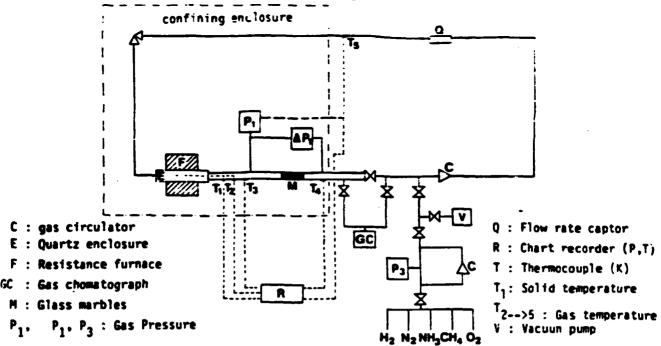
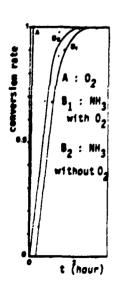
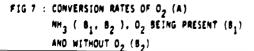


FIG 6 : APPARATUS DIAGRAM

Two qualitative experiments were performed at 973 K. The present gas mixture was  $O_2$ ,  $H_2$ ,  $NH_3$ ,  $CH_4$ ,  $N_2$  in the first experiment, while there was no oxygen in the second one. The initial ratios of gas moles number to uranium surface were respectively  $60 \text{ moles/m}^2$  and  $100 \text{ moles/m}^2$ .

Results: Oxygen was immediately consumed. Then a first part of ammonia reacted with uranium giving a nitride (Fig 7), the other part being decomposed into gaseous nitrogen and hydrogen. We also observed methane and nitrogen reactions with uranium (Fig 8), that are much slower than the first ones. Initially present hydrogen did not react chemically, and ammonia and methane crackings provided the predicted hydrogen amounts.





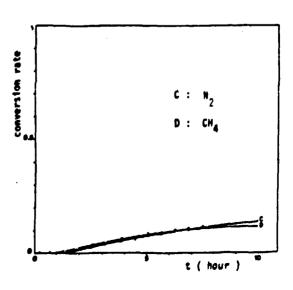


FIG 8 : CONVERSION RATES OF N2 AND CH4

The obvious selectivity of the chemical reactions between uranium and the gaseous mixture, oxygen reactivity being higher than associate then nitrogen and methane cnes, leads to interferences into the system. Oxygen delays associate reaction (Fig. 7) as well as associated delays methane reaction (Fig. 8), while associated decomposition into gaseous nitrogen maskes initial nitrogen conversion rate (Fig. 8). In order to define the influence of the different product layers, progressively formed at uranium surface, on the gas reactivity, we aim to first study each chemical reaction at 973 K, and then to compare the rate constants to their values in gaseous mixture.

### 5.- CONCLUSION.

Qualitative experiments seem to confirm that uranium could be used for hydrogen purification, at 973 K. Our aim is now to study each chemical reaction at different pressures, to predict the reactivity of the gaseous mixture on uranium as a function of its composition. This will let to predict the uranium amount required for the treatment of various composition gaseous mixtures.

### 6.- REACTION OF PURE GAS WITH URANIUM AT 973 K.

We studied each reaction of pure gas with massive uranium at 973 K. In every case, the partial pressure of each gas decreases as a function of time, according to two phases. Experiments show that the first one is governed by a surface kinetic law:  $\frac{dP}{dt} = k S P^n$ , while after a short transition time, gas diffusion into the solid products plays and becomes the limiting step.

### 6.1.- Uranium oxidation by dry oxygen.

Oxygen has completely converted uranium into  $\rm U_3O_8$  powder, the product composition having been analysed by X ray diffraction. Oxygen pressure variation plotting as a function of time, points out the two phases that take place during the reaction.

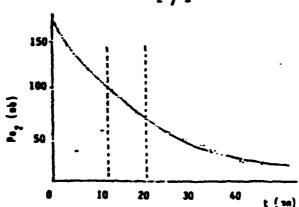


Fig. 9 :  $P_{0_2}$  as a function of time

The initial order of the chemica! reaction is defined as :

$$v_0 = \left| \frac{dP}{dt} \right|_{t=0} = kSP_0^{n_0}$$
. Then  $n_0 = \frac{1}{2}$  (Fig 10)

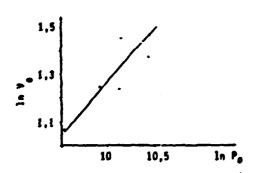
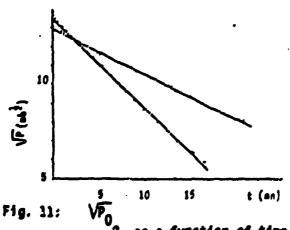


Fig. 10 : In  $V_0$  as a function of In  $P_0$ 

According to the bibliographic studies, we have extrapolated this result to the current order n so that  $n=\frac{1}{2}$ . Figure 11 seems to confirm  $\frac{dP}{dt}=kSP^{\frac{1}{2}}$  with  $k=142\ Pa^{\frac{1}{2}}/s.m^2$ , and S being the initial area of the metal. Hence the first phase corresponds to a linear kinetic law at variable pressure.



e as a function of time, for two values of S

As soon as the oxide layer is thick enough, diffusion through the solid products is assumed to be the limiting step. Experimental points in fact follow a diffusional model ( Fig. 12 ) that let us estimate the effective diffusivity in the oxide,  $D=5.8\ 10^{-6}\ m^2/s$  ( Annexe 1 ). We can then check our hypothesis by comparing the diffusional molar flow of  $O_D$  ( ND ) to the linear kinetic one (  $N_R$  ). In the example of figure 9,  $N_D=4.5\ 10^{-4}$  mole  $O_2$  / s.m² and  $N_R=2.5\ 10^{-3}$  mole  $O_2/s.m²$  at t = 30mn. So, chemical resistance may be neglected before the diffusional one, and diffusion is the limiting step of the second phase.

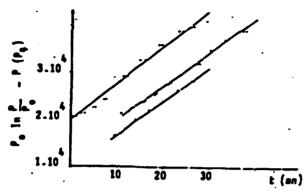


Fig. 12:  $P_0$  in  $\frac{P}{P_0}$  - P as a function of time for  $P = P_{0_2}$  and differents S

As self-diffusivity of  $0_2$  in  $U0_2$  is ranging from  $10^{-11}$  m<sup>2</sup>/s to  $10^{-17}$  m<sup>2</sup>/s at 973 parameters to the oxide stoechiometry  $U0_{2+x}$  [7], the larger value of D=1.8  $10^{-6}$  m<sup>2</sup>/s is likely to correspond to gas diffusivity through the porous oxide  $U_30_8$ .

### 6.2.- Uranium-methane reaction.

We have obtained a solid product shell, thightly adherent to the uranium surface, but we have not been able to define its composition. From the bibliographic investigations we can suppose that we have formed UC or UC<sub>2</sub> and :  $U + CH_4 --> UC + 2 H_2$  or  $U + 2 CH_4 --> UC_2 + 4 H_2$ . Hydrogen has been produced according to the amounts predicted by the chemical reaction (Fig. 13).

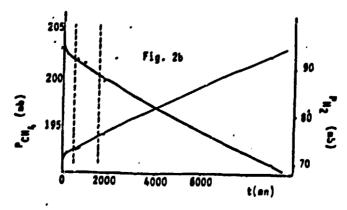


Fig. 13:  $P_{CH_4}$  and  $P_{H_2}$  as a function of time

The initial order of the reaction is  $n_0 = 1$  and we have extrapolated this value to the current order n. The experiment results confirm however that the first phase follows a linear kinetic law:  $\frac{dP}{dt} = k S P$  (S: initial area of the metal) (Fig 14).

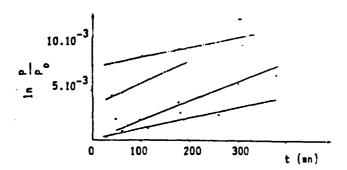


Fig. 14: 1 n  $\frac{P}{Po}$  as a function of time for P =  $P_{CH_4}$ 

Hydrogen does neither perturb the initial kinetic constant  $k_0$ , nor the current value k. But  $k=2.5\ 10^{-4}\ s^{-1}\ m^{-2}$  is for more lower than  $k_0=10^{-2}\ s^{-1}\ m^{-2}$ . This likely implies a deficiency of the interface regeneration due to slow solid diffusion rates. This first phase is a very short one and as soon as the metal surface is covered by a thin carbide film, diffusion into the solid products plays and becomes the limiting step.  $D=1.5\ 10^{-11}\ m^2/s$  is the experimental diffusivity value. Self-diffusivity of carbon and uranium in uranium carbide being respectively  $D_c=9\ 10^{-14}\ m^2/s$  and  $D_u=7\ 10^{-17}\ m^2/s$  at  $T=1500K\ |10|$ , our value does probably not correspond to a chemical diffusion, but to an effective diffusivity in a not very porous solid.

### 6.3.- Uranium nitrogen reaction.

We have obtained two sorts of product layers: a compact shell adherent to the metal surface and nitride powder similar to a mixture of  $UN_2 - U_2N_3$ . The initial order of the reaction is  $n_0 = \frac{1}{2}$  that corresponds to a dissociative adsorption of nitrogen.  $\frac{dP}{dt} = k S P^{\frac{1}{2}}$  defines the kinetic linear law with  $k = 0.4 Pa^{\frac{1}{2}} / s m^2$  (Fig 15).

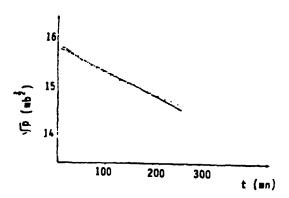


Fig. 15:  $\sqrt{P}_{N_2}$  as a function of time.

As for the methane-uranium reaction we have noticed a break between  $k_0 = 1.3 \text{ Pa}^{\frac{1}{2}}/\text{s.m}^2$  and  $k = 0.4 \text{ Pa}^{1/2}/\text{s.m}^2$ . The effective diffusivity we have measured is  $D = 2.5 \cdot 10^{-10} \text{ m}^2/\text{s}$  and surely represents diffusion in the compact solid.

### 6.4.- Uranium-ammonia reaction.

Ammonia is consumed by two reactions occurring simultaneously: a nitride formation and a catalytic decomposition into gaseous nitrogen and hydrogen. When the nitride layer is thick enough, the whole of the gas is decomposed instead of diffusing to uranium surface. Then nitride is formed according to an only phase: a chemical kinetic one. The platting of  $V = \frac{dP}{dt}$  as a function of proves that each reaction follows a kinetic linear law  $\frac{dP}{dt} = k$  S P with k = 2.5 s<sup>-1</sup> m<sup>-2</sup> for the catalytic decomposition and k = 0.5 s<sup>-1</sup> m<sup>-2</sup> for the nitride formation (Fig 16). Theese two values are independant of  $H_2$  and  $N_2$  content in the gaseous mixture.

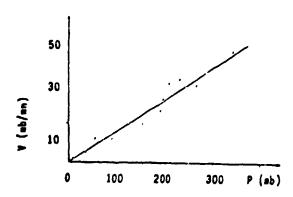


Fig. 16 A : V as a function of  $P_{N/3}$  (nitride formation)

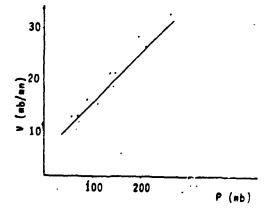


Fig. 16 B : V as function of  $P_{NH_3}$  (Catalytic decomposition)

So, about 80% of NH $_3$  is decomposed into N $_2$  and H $_2$  and the remaining gas forms a nitride with uranium. In this latter case, we have not observed any break between k $_0$  and k. If we admit that nitride formed from ammonia has the same structure as the nitrogen one, this result might imply that NH $_3$  enters uranium more deeply than N $_2$ .

### 7.- REACTION OF GASEOUS MIXTURE WITH URANIUM AT 973 K.

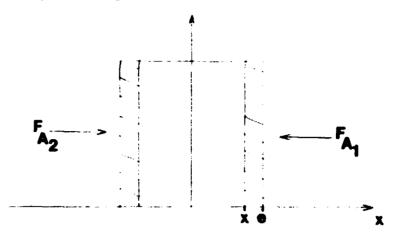
When the gases are mixed all together,  $H_2$ ,  $N_2$ ,  $O_2$ ,  $CH_4$ ,  $HH_3$ , chemical reactions likewise run according to two phases. But we have observed interferences in the system uranium-gaseous mixture. Water formation from  $H_2$  and  $O_2$  acts as a catalyst on the oxidation reaction and enhances the kinetic constant k from 142  $Pa^{\frac{1}{2}}$  /s.m<sup>2</sup> to higher values that depend on the water content ( we have obtained 430  $Pa^{\frac{1}{2}}$  / s.m<sup>2</sup> and 737  $Pa^{\frac{1}{2}}$  / s.m<sup>2</sup>). But a thin oxide layer on the metal surface does not affect the current kinetic constants of  $N_2$ ,  $NH_3$  and  $CH_4$ . If nitride and carbide formation does occur simultaneously, then the experimental current kinetic constant of H<sub>2</sub> and CH<sub>4</sub> are respectively increased from  $k = 0.4 \text{ Pa}^{\frac{1}{2}}/\text{s.m}^2$ to  $k = 1.5 \text{ Pa}^{\frac{1}{2}}/\text{s.m}^2$  and from  $k = 2.5 \cdot 10^{-4} \text{ s}^{-1}\text{m}^{-2}$  to  $k = 1.10^{-2} \text{ s}^{-1}\text{m}^{-2}$ . As 1.5  $Pa^{\frac{1}{2}} s^{-1}m^{-2}$  and 1.10<sup>-2</sup>  $s^{-1}m^{-2}$  corresponds to the initial kinetic constants  $k_0$  of  $N_2$  and  $CH_4$ , we can reasonably suppose that the reactional interface regeneration has been improved in consequence of solid structure defects. Simultaneous formation of nitride and carbide would have promoted solid diffusion rates of N and C (when they curbed the kinetic rates in the case of pure gas (cf 6) ). On the other hand no similar effect has been observed on nitride formation rate from ammonia; but we had pointed out that there was no break between k and ko (cf 6). Nevertheless, the kinetic constant of ammonia catalytic decomposition is reduced from  $k=2.5 \text{ s}^{-1}\text{m}^{-2}$  to  $k=0.2 \text{ s}^{-1}\text{m}^{-2}$ . This latter value being obtained with and without 02, we suppose that CH4 alone has an influence. Finally, hydrogen does not perturb the different gases conversion rates. Theese results point out the importance of the solid surface state on the chemical kinetic constant, but also on the durability of this phase. In fact, experiments have shown that the "limiting thickness" of nitride and carbide layers was greater than with pure gases, so that the step controlled by diffusion into the solid products was postponed. But we have to further study this type of effect in Order to provide accurate quantitative results.

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#### ANNEXE 1

Gaseous diffusion in the product layers



In the case of a massive uranium flat, we can define its initial tickness, e at t = 0, and its thickness x at t, and the solid product thickness e-x at t.  $F_A = F_{A_1} + F_{A_2}$  is the diffusion flow:  $F_A = -2$  DS  $\frac{dc}{dx}$ , c being the gas concentration.

We can neglect gas accumulation in the solid with regard to the gas amount in the closed loop, and hence we suppose a quasi steady state of diffusion.

Then  $F_A$  has a constant value and  $F_A = -2$  DS  $\frac{c(e)-c(x)}{e-x}$  If diffusion is the limiting step, then c(x) = 0, and  $c(e) = \frac{P}{RT}$ .

On the other hand, e - x is proportionnal to the pressure variation in the loop and e - x = a ( $P - P_0$ ),  $P_0$  being the initial pressure, and "a" a constant that only depends of our experimental system.

Finally, in the case of a diffusion limiting step,  $F_A$  corresponds to the molar flow rate of gas consumed by uranium :  $F_A = \frac{V}{RT} \cdot \frac{dP}{dt}$ .

Hence 
$$\frac{dP}{dt} = -b. D. \frac{p}{p_0-p} = > \int_{1}^{T} \frac{p_0}{p} - 1 \cdot dF = \int_{1}^{T} b.dt$$

 $\mathbf{t_1}$  corresponding at the beginning of the diffusion step.

We obtain the diffusion equation: Po Ln  $\frac{P}{Po}$  - P = b D t + c, b and c being integration constants that only depends on the experimental system.

### TASK ACTION T 5 B

## ELEMENTS FOR TRITIUM RECOVERY FROM THE FUSION REACTOR CERAMIC BLANKET

M. ROSTAING

#### TASK ACTION T 5 B

### ELEMENTS FOR TRITIUM RECOVERY FROM THE PUSION REACTOR CERAMIC BLANKET

#### **ABSTRACT**

### ELEMENTS FOR TRITIUM RECOVERY FROM THE FUSION REACTOR CERAMIC BLANKET

We have been considering the case of T2 recovery through a low pressure helium separate circuit for production of INTOR (50 g tritium per day) and NET (91.2 g tritium per day), the partial pressure of T2 in helium being set at 0.1 Pa.

After evaluation of the amount of energy necessary in an INTOR type diagram with T20 trapping in a cold trap that is cooled with liquid nitrogen, cooling has been suggested by work producing expansion of the Helium flow itself, hence reducing the energy requirements to 0.8 MW for INTOR and to 1.5 MW for NET. These figures can be reduced if one considers the possibility of a sweeping circuit under a pressure lower than atmospheric pressure.

Taking into account the dilution with 100 to 1000 H/T which appears necessary in the T2 in-pile extraction experiments, one may condider the application of molecular sieves to the extraction of T20, thus enabling an energy consumption level lower than those involved by the cold traps:

It could be observed that tritium oxidation by means of an excess of oxygen resulted in energy requirements that were almost doubled if cold traps were used whereas these needs were only slightly increased with molecular sieves.

An estimation has been made of the reconcentration of the product diluted with 100 or 1000 H/T based on cryogenic destillation.

The feasibility of the different systems will be established only once experiments have been performed: problem of the frost deposit in the cold traps, molecular sieves 'efficiency on hydrogen and tritium, extraction efficiency when system is operated under a pressure lower than atmospheric pressure, and finally evaluation of the energy drop throughout the extremely complex circuit of the sweeping gas in the blanket.

### CONTENTS

1 - Evaluation of the system energy requirements
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2 - Influence of the dilution with 100-1000 H/T
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### ELEMENTS FOR TRITIUM RECOVERY FROM THE FUSION REACTOR CERAMIC BLANKET

### M. ROSTAING

### ELEMENTS FOR TRITIUM RECOVERY FROM THE FUSION REACTOR BLANKET

We have been considering the case of T2 recovery through a low pressure helium separate circuit for production amounts in the range of those of INTOR (50 g tritium per day) and NET (91.2 g tritium per day). As it is usually the case, the partial pressure of T2 in helium has been set at 0.1 Pa so as to limit the tritium inventory and its permeation.

The recovery circuit for the tritium produced in the blanket is often designed in such a way that tritium becomes oxidized and the tritiated water becomes trapped in a cold trap that is cooled with liquid nitrogen under a 10 bars pressure as it is the case in the INTOR Project. We have estimated the amount of energy required by such a circuit and those required by the corresponding circuit in NET and we have suggested a number of improvements, as for example the use of the helium work producing expansion itself to generate the frigorific power which is required for cooling the sweeping gas and for the condensation of the water.

Now, we know that condensation produces frost that will not deposit easily on the exchanger walls. This frost will have to be retained by means of filters; the carrier gas must then slowly flow through the filters which are placed immediately after the cryocondenser.

The power in the low pressure (\* 1bar) recovery circuit is estimated to be 0.8 MW for INTOR and 1.5 MW for NET.

In order to reduce the permeation by swamping effect and to increase the amount of recovered tritiated water a dilution had been suggested with 100 or 1000 part of H/T. Without such a dilution it would have been necessary, for example in the case of INTOK, to have trapping cycles of 7 days to recover a handable quantity of 1.2 liter of pure tritiated water and as a consequence there were parts with a high inventory content of T2. The in-pile experiments concerning the extraction of T2 from ceramics have shown that this dilution was also highly favorable for the release of tritium. One will have to take this new data into consideration. Although it does not allow to clear all uncertainties concerning the trapping of the frost by cold traps, it will make this trappping easier. The dilution with 1000 H/T enables to consider the potential use of molecular sieves. It was

not possible to use molecular sieves when recovering pure T2 under the form of T20 at 1 ppm in the sweeping gas, the efficiencies of molecular sieves are not well known for this concentration and they are anyway much lower than in the 100-1000 ppm concentration range. In addition the pure tritiated water would have replaced the water irreversibly linked to the molecular sieve's structure and this would have caused an inadmissible loss. A regenerating helium flow which is smaller than the sweeping gas flow enables to recover the tritiated water that has been trapped in the molecular sieves and to regenerate these sieves. The helium flow deposits the recovered water in a cold trap. Cooling of the cold trap can be ensured by the expansion of the regenerating helium flow. The helium flow is brought to a low temperature, therefore it is dry enough to regenerate the molecular sieve in good conditions.

In this system the amount of energy required is considerably reduced: the main flow of the sweeping gas requires only the energy for the circulation of the blower (compression between 1 and 2 bars) and only the regeneration mass flow is compressed in order to generate the cold power required by the cold trap, for a fraction of the day only.

If, as it has been suggested, the tritium oxidation must be made with an excess of oxygen, elimination of the oxygen in excess must occur in a second trapping unit for the H2O in excess, thus the energy requirements of the system with cold trap is almost

doubled whereas the energy consumption of the recovery system with molecular sieves is only slightly increased.

A sweeping system has also been considered with a reference pressure lower than atmospheric pressure, such a system would appreciably reduce the energy requirements and the investments, more particularly when molecular sieves are used.

And finally, we have made an estimation of the size of the reconcentration columns for the product diluted with 100 or 1000 H/T, they should be added to the isotopic separation system.

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1 - EVALUATION OF THE SYSTEM EMERGY REQUIREMENTS

1 - 1 The case of INTOR

Figure 1 reminds of the main characteristics of the reference

circuit (1): compression of the sweeping gas under 10 bars for

trapping operations in the cryocondenser and production of liquid

nitrogen (N2L) which is used in the cryocondenser to cool the

gas.

Compression : cf.Appendix 1

" Gas flow rate: 7800 Nm3/h

Compressor power : 814 kW

Frigorific power: the 402 kW which are required for cooling

helium are obtained from the latent heat of the nitrogen which

evaporates at 77 K (-196°).

Nitrogen requirements :

402 kW

= 2 kg/s = 7200 kg/hour

latent heat N2 : 198 kJ/kg

Power of the N2L frigorific system : since the saturating vapor

of the nitrogen can be recovered, it is not necessary to cool

nitrogen from ambiant temperature down to the point of liquefac-

tion: 225 kJ/kg.

As :

Theoretical power

198 + 225

### Total required power

Pc + Pf = 2696 kw

for NET it corresponds in the production ratio of T2, to

50 g/j

### 1 - 2 Possible improvements

It has been proved that (2) filters were necessary to stop the fog of ice cristals which forms in helium while it is cooling, and the probability that there would not be any deposit on the CT cryocondenser walls has ben confirmed by (4). One could retain the scheme on Figure 2, in which helium is pre-cooled before flowing into the cryocondensor, down to a temperature somewhat higher han the T20 dew point, this means - 50 ° as the temperature at the cryocondenser entry. In other respects an expansion turbine on the helium that flows back to the blanket (under 2 absolute bars) would have enabled to recover a large fraction of the compression energy (Appendix 1).

The energy needs would be reduced to

(Pc = 814 kW) - (P turbine = 301) = 513 kW

Pf for 270 kW at tN2L : 1264 kW

Total absorbed power INTOR: 1777 kW

Total absorbed power NET : 3241 kW

NOTE: for power reactors (450 g/j T2) 16 MW

### 1 - 3 Refrigerating cycle with sweeping gas

The two following points have been underlined:

- a) Recovery of the frigories contained in the helium which flows out of the cryocondenser is not feasible in the INTOR main refrigerating system since the N2 cycle works with saturating vapors recovery, frigories are not needed above 90 K.
- b) As the tritium to be recovered is diluted with 1000 or 100 volumes of H2, it is not possible to use an exchanger of type E2, (Figure 2), anyway the higher dew points let expect a less intense cooling of helium in the CT.

Dew points	Dilution	
<b>!</b>	100 H/T	1 1000 H/T
at p = 1 bar	-42 °C	-20 °C
p = 2 bars	-35	- 12
p = 10 bars	-20	+ 7

At -120 °C the H20 partial pressure is  $\approx 10^{-6}$  Pa. We could choose this temperature for the CT trapping, provided that the frost filters prove to be efficient.

If the helium flowing out of CT under a 10 bars pressure at 150 K is submitted to an expansion under 2 bars (Cf. Appendix 1-3), then the helium will have a temperature of 97 K and be able to cool the sweeping gas in the CT tubular beam (Fig 3), the characteristics of which are given in Appendix 2, with a 310 kW power, out of which 301.2 kW are used for helium cooling and 4.8 kW for water condensation in the case of a dilution with 1000 H/T and for INTOR. For NET total power reaches 565 kW for exchange.

The compression of the gas enables to cover the total energy needs of the circuit , cf.1-1, for INTOR : 814 kW

for NET : 1424 kW

NOTE: in this new layout, cooling of helium between oxidation and compressor must occur by means of cooling water.

### 1 - 4 Sweeping circuit with pressure under 1 absolute bar

A system with reduced pressure at the blanket outlet would permit to reduce in proportion the mass flow of the helium that carries tritium with a pressure set at 0.1 Pa. But one is then faced to the following drawback: there must be a turbulent Reynolds number for the heat exchanges.

The critical point is the high temperature helium exchanger at the blanket outlet in which there must be a turbulent Reynolds number:

#### with Re > 2500

v : gas velocity, we could take 100 m/s

d : tube diameter; it should not vary much from the 6 cm of INTOR. This value together with v = 100 m/s results in exchangers very "extended" in length and in a small number of tubes.

C: Specific weight of the gas

 $Q = Co \times P'$  Co = minimum value of at 1 bar and at highest temperature of the system (blanket outlet) = 0.058 kg/m<sup>3</sup>

P' (bar) : operating pressure considered

 $\mu$  : gas viscosity, critical value is  $\mu$ , which is the highest

value of the system (at blanket outlet temperature) :
3.9 10 Pl.

$$2500 \times 3.9 \quad 10^{-5}$$
 Which gives : P' >  $\frac{100 \times 0.06 \times 0.058}{100 \times 0.06 \times 0.058}$ 

#### P' > 0.284 bar

With P' = 0.3 bar, the computation leads in the case of INTOR to high temperature exchangers (there are 12 of them, one for each module) composed of two tubes with diameter 6cm and length 10.2 m because of the gas high velocity. For the "power reactor" size, we would have 18 tubes of 10.2 m in parallel for one module in the case of a 12 modules design. The sweeping gas at 0.3 bar (= 30 000 Pa) and ambient temperature contains the tritium at 0.1 Pa, diluted by H2 at 100 Pa. After oxidation and compression with a factor 10, the partial water pressure in the gas is 1000 Pa with a dew point of + 7 °C.

Trapping at - 73 ° (partial pressure of H2O = 0.25 Pa) is

deemed to be sufficient. Expansion of the gas from 3 to 1.4 bars enables to have 163 K instead of 200 K for the cold traps self-cooling (cf. Appendix 1-4). In that case the compression energy needs are only 30 % of those of a system with a 1 bar initial pressure, i.e. 250 kW for INTOR and 456 kW for NET, however solely where there is a dilution with 1000 volumes of hydrogen and provided that experimental results of trapping at 200 K are available.

### 2 - INFLUENCE OF THE DILUTION WITH 100 - 1000 H/T

The dilution with 100 to 1000 volumes of H2 (or D2) for one volume of T2, offers, in addition to the favorable consequence for the tritium release which could be observed with the in-pile experiments, the advantage of considerably reducing the tritium permeation more particularly to the helium (or water) cooling system and in other respects it enables to recover in the course of trapping operations more practical quantities than in the case of pure T20 recovery as for example in INTOR where 7 days cycles were necessary; this had the evil effect of creating a very important inventory point of T2. In other respects, with the dilution we are again in the concentration areas proper to the use

### TABLE 1

### DILUTIONS WITH 100 AMD 1000 H/T

Amounts to be recov T2 g/j T20 "	ered	INTOR 50 183	NE 9 33	11.2
HE sweeping N.m /h		7 800	14 20	00
Dilution by 100				
H2 m <sup>3</sup> /h HTO + H20	kg/j	0.78 15.2	ž	1.42 ?7.3
<b>Dew</b> point	at 1 bar at 10 bars		- 42 °C - 20 °C	
Dilution by 1000				
H2 m³/h HTO + H20	kg/j	7.8 150	2	14.2 ?70
Dew point	at 1 bar at 10 bars		- 20 °C + 7 °C	

of molecular sieves. The He-hydrogene separation under the form of H2,HT has not been considered here. Indeed, because of the swamping effect a separation by means of permeation membranes is not possible, it would result in a too much diluted product, and the use of getters is not adapted to the treatment of very important flows: cf.appendix 3.

With the 1000 H/T dilution the release of H20, HTO formation heat is no longer negligible, the sweeping gas temperature increases by 11 °.

The values obtained with the hydrogenous species being oxidized on a stoechiometric way by 02 are given in Table 1. The consequences of using an excess of oxygen as recommended in (8) will be studied in the last part of this chapter.

### 2 - 1 Cryogenic trapping

The recorded dew points allow to consider trapping either by cooling cycle by the sweeping gas itself, cf.Appendix 1, after expansion the temperature is then 128 K, or by means of a freon frigorific machine (level around - 70 °C), should the experience prove that collection of the frost can be efficiently performed under these conditions: cryogeny specialists of the S.B.T.(Low Temperature Division at C.E.N.G. Grenoble) believe that this pilote scale experimentation is absolutely necessary as a preliminary to any cryocondenser project.

Compression under 10 bars is no longer justified as it was the case in the former concept without dilution and with increased partial pressure of T20, however compression under 10 bars enables using the self-cooling cycle, reducing the size of the device and easily obtaining low velocities (0.1 m/s) in the frost traps which are placed in the lower part of the cryocondenser.

The volume of water to be recovered enables to consider cycles of one day or even less, hence considerably reducing the inventory at this step of the circuit, which had a strong influence on the total tritium inventory (3).

On the other hand we are facing the problem of the accumulation as frost in the trap, in the filters, and also possibly in a number of the exchanger tubes, should the distribution of gas flows be in disequilibrium. 150 kg of frost (case of INTOR, one day cycle) may occupy up to more than one cubic meter depending upon the cristallization and deposition conditions which can be precisely defined solely by experimentation.

### 2 - 2 Molecular sieves

The use of molecular sieves to trap pure T20 was hindered by the low proportion of T20 in helium under partial pressures (0.1 to 1 Pa) which were outside the range of the results already experien

ced with the molecular sieves. In addition the residual water content could result in an inadmissible irreversible trapping of tritiated water, and to have a low residual inventory one should have used very high regeneration temperatures, hence reducing the time life of the molecular sieves and the desorption flow rate (helium) would have been of the same magnitude as the sweeping flow rate (8) and consequently there would not be any interest in using molecular sieves since the final trapping of T20 is necessarily made by means of cryogenic traps on the desorption helium.

With dilution from 100 to 1000 H/T we are again in the already experience: range of use and the tritium concentration in the irreversibly linked water being in inverse ratio to the dilution, the T2 fixation is no longer prohibitive (provided that there is no preferential trapping effect on T2).

2 - 2 - 1 Trapping on molecular sieves (Figure 4) could be realized under two absolute bars at the circulator strain (1 bar absorbed at N.P.T. aspiration + 1 bar lost in the circuit) which considerably reduce the energy consumption for circulation, to 240 kW for INTOR and to 440 kW for NET.

As regards the specifications of NET (1000 H/T), C.E.C.A. was kind enough to give us the following dimensions and conditions:

### A - Basic data for computation

. Flow rate (MM <sup>3</sup> /H)	14 000
. Type of supply	helium
. Pressure (abs.bar)	2
. Temperature (degree C)	25
. Phase	gas
. Water content (PPM V)	1000

### DIMENSIONS

### B - Adsorbtion

### . Type of adsorbing agent :

### Molecular sieves CECCA

siliporous	SBF - 426 -SG
. Columns	2
. MS quantity per column (kg)	2360
. Internal diameter (mm)	1500
. Height of MS coating (mm)	1930
. Adsorption time (hours)	24
. Direction of flow	from top to bottom
. pressure loss (bar)	0.2

#### C - Regeneration

#### C1 - Heating :

. Flow rate (MM <sup>3</sup> /H)	2000 helium
. Pressure (abs. bar)	2
. Temperature (degree C)	290
. Time (hours)	10
. Direction of flow	from bottom to top
. Pressure drop (bar)	0.04

#### C2 - cooling

. Flow rate (NM <sup>3</sup> /H )	2000
. Time (hours)	2
. Temperature (degree C)	25
. Direction of flow	from top to bottom
. Pressure drop	0.015

#### D - Note

Water content in the dry product : below 1 ppm V.

Residual water content in the molecular sieve would be 1.5 g per 100 g of sieve, i.e. :

For NET (5.34 g for INTOR).

2 - 2 - 2 The description circuit can be composed of cryocondenser and selfcooling by means of helium expansion (cf.Appendix 1-5 and Figure 4). Its size would be 1/7 of that of a sweeping circuit. However one must take advantage of the higher partial pressure of the water in the description (x) gas than in the sweeping gas. And it is possible to trap 90 % of the water to be collected in liquid form during a first cooling step with cold water supplied by a frigorific set of 37 000 frigories /h
15 kW electric power needs (10). At the exchanger outlet (PT) one must have a cyclon to separate the gas small drops and 9/10 th of

<sup>(</sup>x) CECA indicates 4 % in carrier gas at 2 bars, i.e.

P = 0.08 bar and dew point at 42°C.

the 1 per 1000 tritiated water can be recovered in this way. The rest, under a partial pressure of 0.009 bar corresponding to a temperature of 5°C, will be trapped after it has been compressed at 7 bars (in two compression steps, required power: 102 kW for NET, 56 kW for INTOR, operating 10 hours a day which corresponds to a desorption cycle). The 7 bars pressure is sufficient to ensure cooling temperatures of 175 K ( $\approx$  -100 °C,  $\approx$  2.10 Pa) for the carrier gas and 128 K obtained by expansion at 2.3 bars (Appendix 1 - 5).

NOTE: As to the desorption gas preheating, use of the helium flowing out of the blanket has not been retained because of the tritium permeation risks.

#### **ENERGY CONSUMPTION**

	NET	INTOR
Cryocondenser	1484 kW	814
with helium cycle		
System with molecular sieves		
sweeping circulation	440	240
trapping - cryocondenser (*)	102	56
- auxiliaries (*)	67	37
	609	333

(\*10 hours a day only)

#### 2 - 3 Oxidation with an excess of oxygen

In order to limit the risks of tritium permeation one has to consider a very early oxidation of the hydrogenous species in the extraction system at the blanket outlet, however the hot walls (tubings, high temperature exchangers) may cause a partial reduction; D.LEGER (8) suggested therefore to infuse an excess of oxygen and to have three oxidation steps: (Figure 5):

- one at the blanket outlet,
- a second one at the temperature level of 200 °C ( as in INTOR), below which there is no further reduction to be feared on the Walls.
- and a third oxidation step, following the tritiated water trapping stage, so as to combine the 02 excess with the precise H2 quantity necessary to eliminate

02 and to ensure 1000 v.p.m. of H2 in the sweeping gas on its way back into the blanket, after trapping of the "secondary water" that should not be tritiated, or if so, at a very low level.

The impact of such a system leads to double the trapping devices:

- Cryocondensation (Figure 3) a second complete device (compression, cryocondenser, frigogenous expansion) must be installed on the sweeping circuit. The energy needs are practically doubled,
- Molecular sieves: the second trapping device on the sweeping circuit does not involve any considerable pressure drop and circulation energy increase. Only the power level of the desorption circuit devices have to be doubled.

The example of NET:

with cryocondensation 2890 kW

with molecular sieves 793 kW

3 - RECONCENTRATION OF RITTUM DILUTED WITH 100 OR 1000

WOLUMES OF HYDROGEN IN THE SMEEPING CIRCUIT

After electrolysis and dessication the hydrogen mixture flows to the cryogenic distillation device. As in the ILL facility a column will ensure the following operations:

- a) Stripping of the tritium contained in the dilution hydrogen in the upper part, the stripped hydrogen being recycled to the sweeping circuit (for a first estimation the extraction ratio is 95 %).
- b) Tritium is enriched in the lower part of the column. 50 % HT are retained at the column bottom.

The precise values for stripping and enrichment will be defined within the frame of the optimization of isotopic separation as a whole, that will indicate whether the product at 50 % HT can be considered for isotopic separation or an additional column has to be installed between 50 and 90 % HT as a preliminary condition.

In the stripping section the L/L minimum ratio chosen is 1.5.

Using the Choffe and Gladel formulas (16) a computation program has been established for the number of theoretical stages in both depletion and enrichment sections with the simplifying assumption that the mixtures are binary ones. It is applied and verified with a good approximation in a recent study of Savannah River Plant (17). Reminder Note:  $\alpha$  H2/HT = 1.5,  $\alpha$  D2/DT = 1.2.

#### Columns diameter

They are inferred from the gaseous L+C flow rate in the depletion zone and from the specific flow rates in the experimented filling material: CY Sulzer: 6000 moles/dm<sup>3</sup>.hr, HETP: 11cm and filling material type DIXON for narrow sections: 1700 moles/dm<sup>3</sup>.hr, (HETP: 5 cm). CY is used for large flow rates. The specific flow rates are those of Deuterium. They should be higher for hydrogen (ratio of the √specific weights), but since we have at the column bottom a mixture with 50 % HT we keep the specification of D2.

The results for NET are given on Table 2.

The use of D2 appears as a disadvantage.

A dilution by 1000 instead of 100 involves rather more important columns.

TABLE 2

Case of WET (92.2 g T2 / Day)

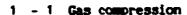
		Dilution 100		Dilution	1000
		Н2	D2	Н2	D2
Flow rate C -	Moles/h	66.5	66.5	665.7	665.7
	N.m³/h	1.49	1.49	14.9	14.9
Flow rate L -	Moles/h	186	465	1913	4736
Flow rate L+C	N.m³/h	5.65	11.9	57.3	121
Number of HETP					
Deple	tion	22.3	41	22	40.5
Enric	hment	12.7	28	18.1	40.2
Total		35	69	40.1	80.7
Height of filling					
materia	1 (m) Cy			4.5	9
D	IXON	1.75	3.45		
Diameter of co	lumns (cm)	4.4	6.4	7.4	9.6
frigorific pow	er (kW)	5.1	14.4	52	147

<sup>(1)</sup> C Feed flow rate, gas

<sup>(2)</sup> L liquid reflux flow

<sup>(3)</sup> L+C Gas flow in stripping section

#### APPENDIX 1



Compressors available at the moment for flow rates of 400 to 700 g/s of He are based on a screw and oil system. These compressors cannot be used with tritium. We have made an estimation of the power of a compressor which would be well adapted to the require ments of tritium taking the assumption of a device that functions as an alternative three stages compressor each stage having a compression rate of  $\sqrt{10} = 2.154$ , the device is assumed to have a yield 7 of 80 % as compared to the adiabatic cycle.

power of one stage = n RT 
$$\frac{y}{y-1} \left[ \binom{p_1}{p}^{\frac{y-1}{y}} - 1 \right] \frac{1}{7}$$

and Pc = 814 kW for INTOR

1484 kW for NET

#### 1 - 2 Expansion turbine

Provided that a machine capable of recovering mechanical energy should be feasible, it is possible following the diagram of Figure A-1-a to heat the gas under 10 bars up to a temperature of 425

K (with a reverse current with the hot helium flowing out of the blanket), characteristic point is A. An isentropic expansion would follow segment AB and we would have the following maximum enthalpy variation:

$$h_A - h_R = 1037 \text{ kJ/kg}$$

Assuming that the turbine yield is 75 %, the variation of practical enthalpy can be determined:

$$h_A - h_C = 1037 \times 0.75 = 777 \text{ kJ/kg}$$

following segment AC (C corresponds to the helium temperature after expansion).

The power produced by the turbine in the case of INTOR (flow rate 0.387 kg/s) is :

$$Pc = 0.387 \times 777 = 301 kW$$

#### 1 - 3 Expansion of the gas to produce the frigorific power

On Figure A-1-b as in the paragraph hereabove, from point A : T = 150 , p = 10 bars one defines point C such as  $h_A - h_C = 0.75$  ( $h_A - h_B$ ) and as a consequence Tc = 97 K.

#### 1 - 4 Case of the system with reduced pressure . Figure A-1-C.

If one wants to work at pressures between 3 bars and 1.4 bar, the expansion to the low temperatures results solely in a small decrease of T. One would have either to spend an additional compression energy, or to have a relatively large surface of exchange of the cryocondenser.

At temperatures around 200 K, the use of which can be considered for dilution with 1000 H/T, the temperature difference obtained is 37 °C.

#### 1 - 5 Description cycle, refrigerated by helium expansion

The diagram A-1-d shows that with pressure at 7 bars and 175 K as the trapping temperature in the CT, it is possible to obtain a temperature or 128 K for the helium in the system, by means of a 2.3 bars expansion.

#### APPENDIX 2

1 - He/He exchanger of the cryocondenser INTOR

t after expansion = 97 K

$$x$$
 - Cooling of the gas  $q_4 = g \times Cp \times \Delta t$ 

g = 0.387 kg/s

Cp = 5.19 kJ/kg

- Cooling of water condensation

$$150 \text{ kg/day} = 0.00173 \text{ kg/s}$$

$$2842 \text{ kJ/kg}$$
  $q_2 = 4.8 \text{ kW}$ 

- Power 
$$P = q_4 + q_7 = 306 \text{ kW}$$

- flow in the tubes. If we adopt the layout of INTOR
with 100 tubes of diameter 0.06 m the Reynold numbers are
relatively high for the frost deposit but we know that
filters are absolutely necessary to trap the frost.

$$Nu = hd/K = 0.023 Re^{0.8} Pr^{0.4}$$

with p = 1 MPa  $tm \approx 225 \text{ K}$  one obtains  $h_{\zeta} = 38.8 \text{ W/m}^2 \text{ K}$ and pressure drop= 900 Pa - Tubes outlet Kern method according to (6)

$$Nu = \frac{h \ d}{k} = 0.36 \ Re^{0.55} \ Pr^{0.33}$$

Outlet temperature = P/gC + 97 K = 249.3 K average temperature 175 K Pressure 200 KPa

One obtains  $h_c = 108 \text{ W/m}^2 \text{ K}$ 

Overall exchange coeficient  $\alpha = 29 \text{ W/m}^2 \text{ K}$ 

Exchange surface =  $\frac{P}{\ll \Delta t_m}$  = 202 m<sup>2</sup>

Tube length = 10.7 m

Calandria section =  $0.85 m^2$ 

Calandria diameter = 1.04 m

Pressure drop = 20 000 Pa

2 - The corresponding beam for NET is extrapolated in proportion to the flow rates

Power rate 565 kW

Number of tubes 182

calandria section 1.55 m²

Calandria diameter 1.4 m

3 - Beam for He/He cryocondenser at 300 kPa/140 kPa with trapping temperature at 200 K and temperature after expansion at 163 K.

Using the same method the following datas are defined for INTOR

Power rate	65.2 kW	
Diameter of tubes	0.02 m	
Number "	164	
Length "	3.7 m	
Overall coefficient	52 W/m² k	
Exchange surface	38 m²	
Pressure drop	580 Pa	
Calandria diameter	0.48 m	

#### APPENDIX 3

#### The use of getters to recover tritium from the sweeping mas

From information data given by specialists (11),(12), it appears that the absorption kinetic is very difficult to define for the reduced pressure, and even more complicated by non uniform phenomena of heat transfer and the thermal conductivity depends from the hydrogen content.

The LASP (Laboratory for Special Aplications of Physics used to take Mg Ni to extract Tritium from the FBR' Sodium (regeneration of the trap) (in preference to Uranium for capacity and kinetic reasons).

The getters' structure is a very delicate one since hydrogen creates an interstitial deformation (12) which has a tendency to lead to a progressive decay after ten or a dozen cycles; their stability is much lower than that of molecular sieves (a few thousands or so cycles and numerous industrial applications).

Most getters do not function satisfactorily in the presence of Helium (14), the SAES getters (15) made of ZrAl are well adapted.

But the slow kinetic of the gas-solid reaction (13) requires slow flow rates, lower than 1 m/s. The velocity of the flow is also limited by the brittleness of the materials which could form dust occlusing the pores.

In Canada (CFFTP) (1%) the use of getters is limited to  $200~\text{m}^3/\text{h}$ , however for higher flow rates they suggest "high towers and fluidized beds", they describe this suggestion as "new and difficult to develop" and wonder whether a fluidized bed made of a brittle material could last a long time.

With the ZrAl, SAES of the larger size (12 kg of ST 101 getters) a mere 10  $\,\mathrm{m}^3/$  h can be processed while in the case of NET there will be 14 000  $\mathrm{m}^3/\mathrm{h}$  with a 0.1 Pa Tritium content to be processed.

As regards uranium the permissible flow rate if 30  $m^2/h$  per bed of 6 kg U.

The use of getters is limited to the low flow rates and to relatively high  $H_2$ ,  $T_2$  partial pressures like in the TSTA.

In other respects the loss of pressure of the SAES elements reaches 1.5 bar.

The following case can be considered: extraction of the tritium permeated to the coolant when the sweeping gas is diluted with

1000 H/T and when the cooling gas contains 60 Pa of H .

Hydrogens to be extracted 133 moles /day = 124 1/hr

The flow rate at 60 bars to be treated 206 m<sup>3</sup>/h

Note: the present SAES elements operate under 5 bars.

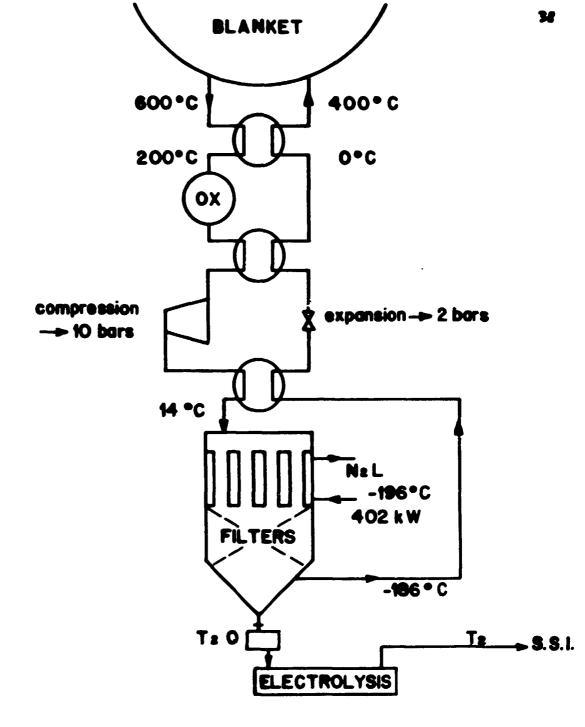
One could consider a range of 21 elements in the absorption phase (and 21 elements in the regeneration phase). The capacity per element being 200 l TPN, saturation will occur after 34 hours, thus we can have a high number of cycles.

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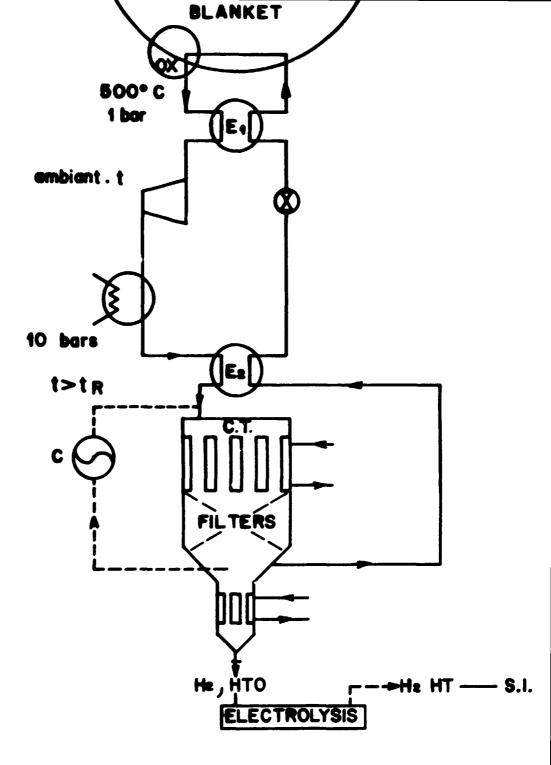
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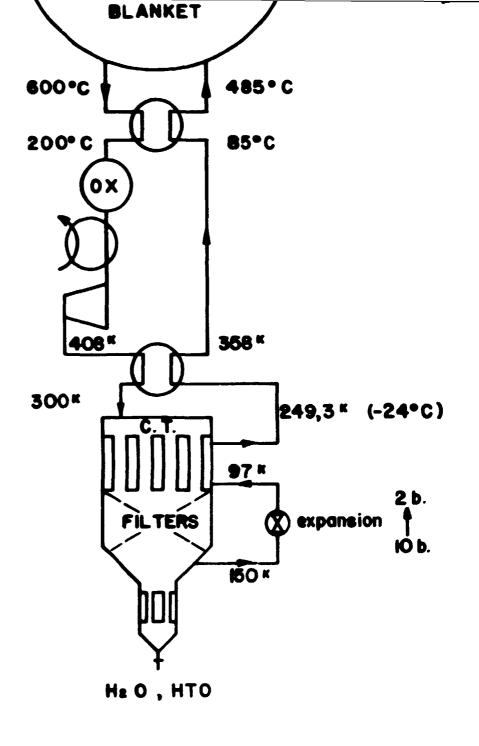
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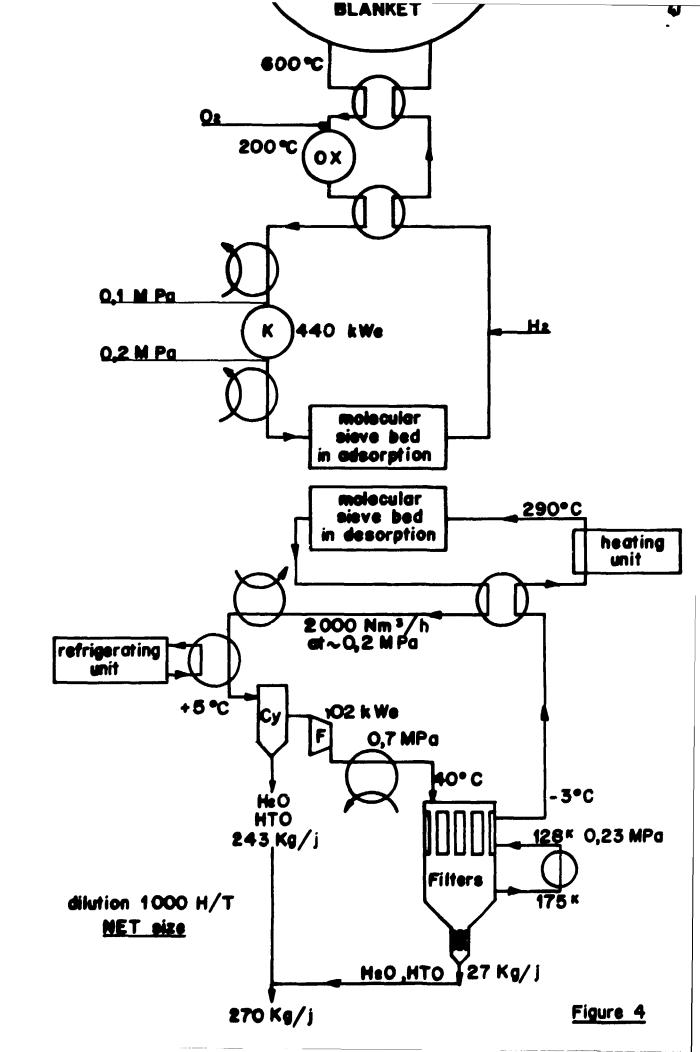
### Schema MTOR

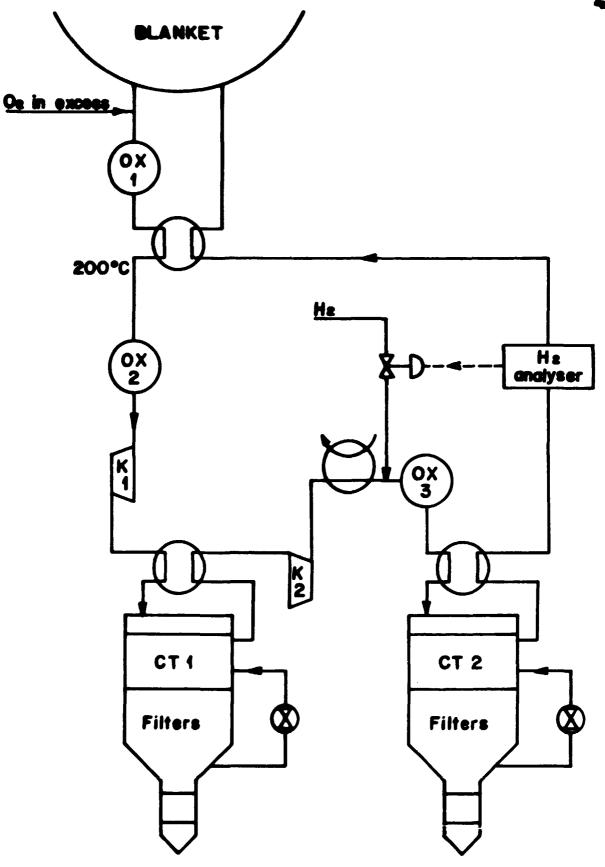
50 g Tz/day
7800 Nm³/h sweeping Helium
He pressure at blanket outlet : 10° Pa
Tz partial pressure at blanket outlet : 0,1 Pa
Δp of circuit 10° Pa

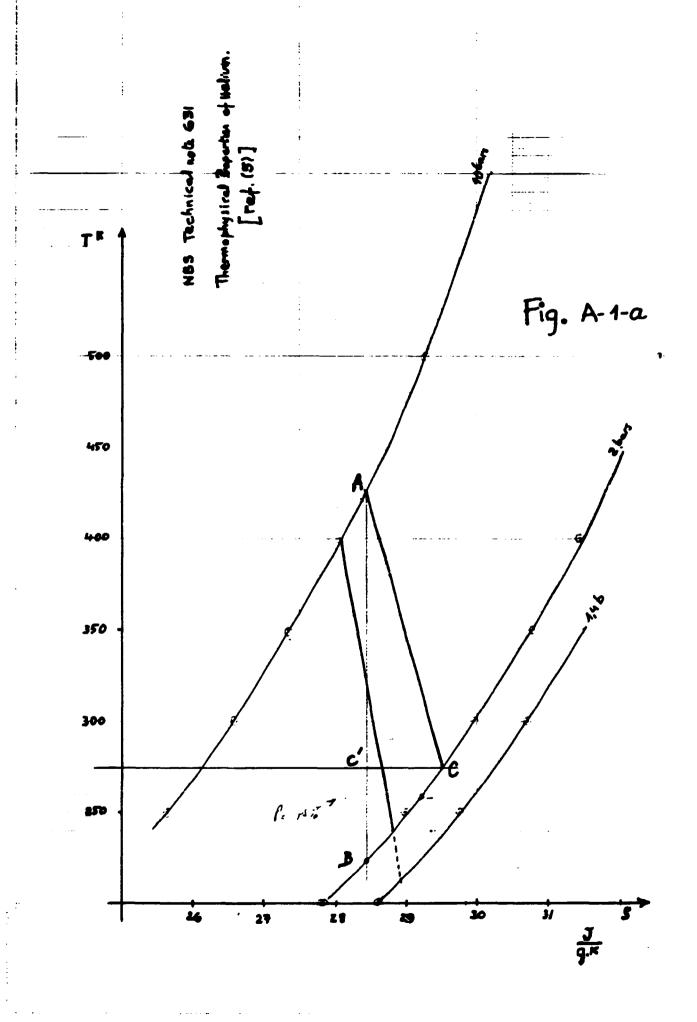


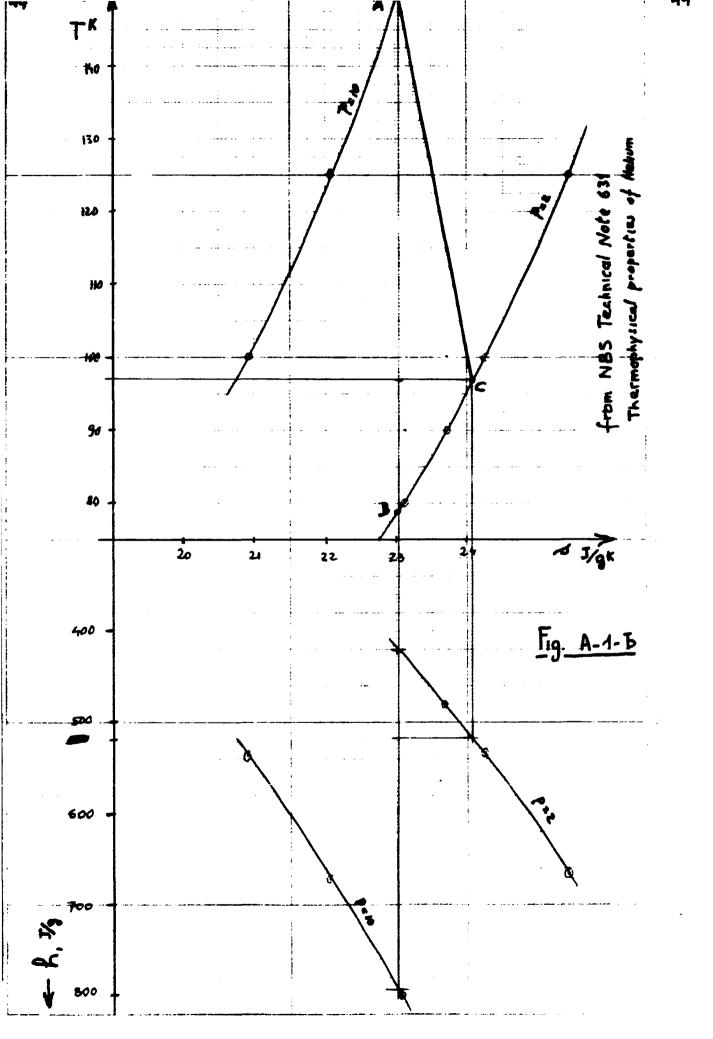


Self -cooled helium loop









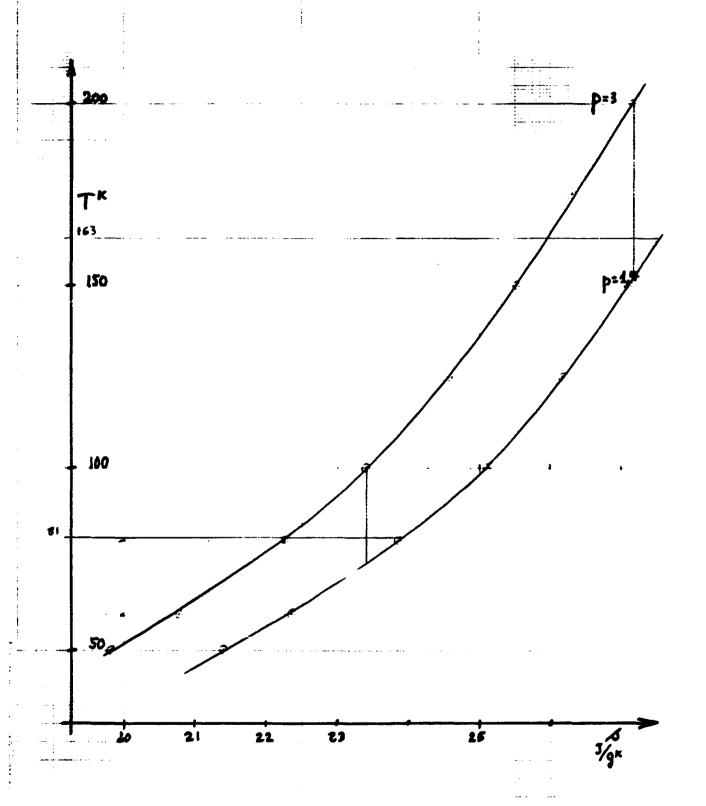
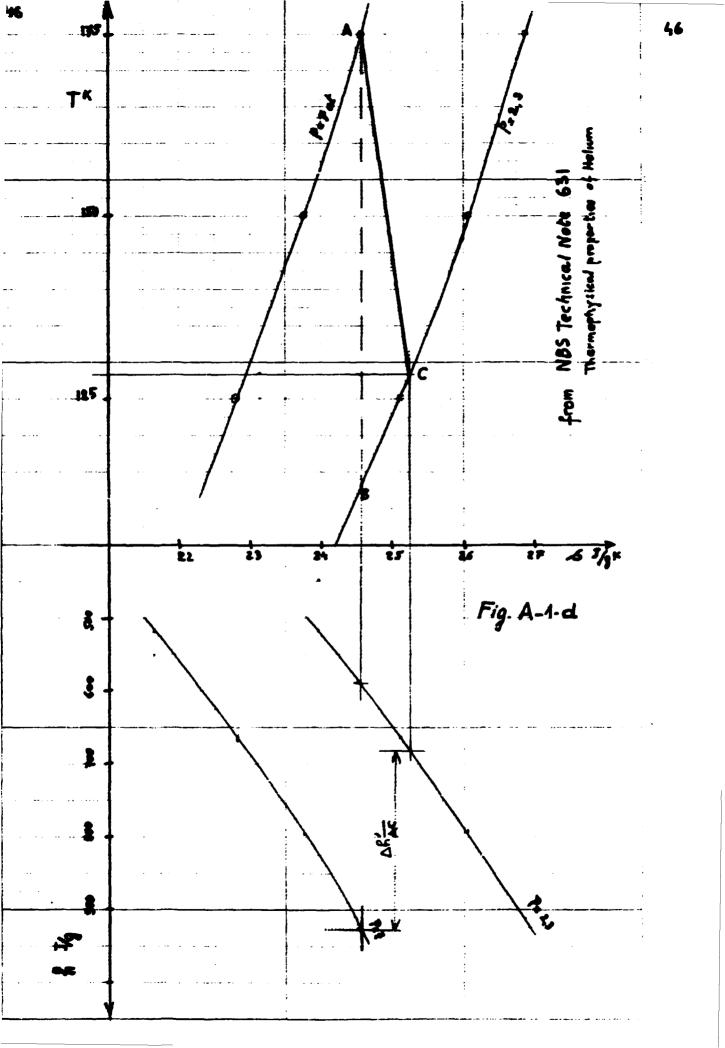


Figure A-1-c



# TASK ACTION T6 LARGE COMPONENTS FOR THE TORUS VACUUM CIRCUITS

C. LORRAIN

#### TASK ACTION TO

# PROGRESS REPORT ON THE T6 TRITIUM TECHNOLOGY TASK (LARGE COMPONENTS FOR THE TORUS VACUUM CIRCUITS) period January 1983 to December 1986

#### **ABSTRACT**

The T6 Tritium Technology contract granted to the CEA by the EEC for the development of large components for the Tritium System corresponded initially to the four following actions:

- 1/ The listing of the components to develop for the realization of the vacuum circuits of the large fusion reactors and their neutral beam injection.
- 2/ Inquiries to determine the environment and operating conditions of these components. Starting form the obtained basic data, elaboration of their individual "Technical Specifications".
- 3/ Assessment of the actual technical rapacity of the European community industry to realize these components.
- 4/ Design, realization and operation of a test loop operated in tritiated media, for intermediary size valves (maximum diameter : 1 m).

During the first step of the study the list of the components has been established:

- gate valves (Torus and Neutral injector)
- fast shutter valve (Neutral injector)
- roughing pumps
- turbomolecular pumps
- cyogenic pumps

Technical specifications and industrial inquiries have bean realized for the Torus gate valves, roughing pumps and turbomolecular pumps according to the priority hierarchy established by the EEC and the NET team.

The industrial inquiries have outlined the necessity of granting feasibility contracts to the European competent industrial corporations. A procedure has been elaborated by the EEC (AHG) and applied by the T6 working group for these contracts, of which two have already been granted and a third one is under procedure:

- granted contracts :
  - PFEIFFER for a turbo molecular pump (50 000 1/s capacity) dated, April 1986
  - VAT for a gate valve (1500 mm diameter) dated March 1987
- contract under procedure :
  - roughing pump

The PFEIFFER feasibility study is progressing according to the programm (12 month study) and the VAT feasibility study is just starting (14 month study).

The initial contract included some investment aimed at hot cell experimentation of prototype valves with tritium for wich an engineering file has been prepared. After a financial (CEA and EEC) and technical analysis (adoption of all metal valves) the realization of any test loop has been postponed.

#### RAPPORT INTERMEDIAIRE, au 1.1.1987, sur le Programme technologique T6

## (DEVELOPPEMENT INDUSTRIEL DES GRANDS COMPOSANTS DESTINES AUX CIRCUITS TRITIUM)

#### C. LORRAIN

Ce programme de Technologie Tritium, confié par la CEE à un Groupe de Travail franco-allemand (GT6) composé de membres du KERNFORSCHUNGSZENTRUM KARLSRHUE (KFk) et du CEA (IRDI/DESICP/DEI) prévoyait les quatre actions suivantes :

- 1/ Etablissement de la liste des composants à développer pour la réalisation des circuits de vide des grands réacteurs de fusion et des dispositifs d'injection de neutres.
- 2/ Réalisation des enquêtes nécessaires pour déterminer l'environnement et les conditions opératoires de ces composants. A partir de ces données de base, établissement de leurs "spécifications techniques" individuelles.
- 3/ Evaluation de la capacité technique actuelle de l'industrie européenne communautaire à réaliser ces composants.
- 4/ Conception, réalisation, exploitation d'un banc d'essais fonctionnant en phases réelles tritiées, pour vannes de dimension intermédiaire (diamètre maximum : 1 m).

Le planning initial de cette action est donné par le tableau joint au texte du contrat (cf annexe).

Au cours des études préliminaires la liste des composants des circuits de vide des réacteurs de fusion a été dressée :

- vannes d'isolement (Tore et Injecteur de Neutres)
- vannes à fermeture rapide (Injecteur)
- pompes primaires
- pompes turbo moléculaires
- pompes cryogéniques

Des spécifications techniques, avec de nombreuses versions (cinq à sept) ont été élaborées et des enquêtes ont été lancées auprès des industriels de la Communauté pour évaluation de la capacité technique actuelle de l'industrie européenne à réaliser ces composants. Elles concernaient les vannes d'isolement des circuits du Tore, les pompes primaires et les pompes turbe moléculaires, conformément à une hiérarchie des priorités établie par la C.E.E. et l'équipe du NET. (NEXT EUROPEAN TORUS)

Les enquêtes industrielles ont mis en évidence la nécessité pour atteindre les objectifs choisis, de lancer auprès de quelques industriels compétents, des contrats de faisabilité, selon la procédure imposée suivante, dont l'application a été confiée aux groupes de travail T6 (GT6 et AHG) \*:

- Etablissement d'une liste des Sociétés communautaires compétentes à consulter (GT6).
- Rédaction par composant d'une "Justification d'une étude de faisabilité" (GT6).
- Rédaction, par composant, d'une spécification technique correspondant au contrat de faisabilité (GT6).
- Etablissement d'une liste réduite des Sociétés retenues pour les appels d'offres (AHG).
- Sélection des propositions (AHG).
- Lancement du contrat commercial d'études (CEE).
- Suivi de l'étude (AHG) (GT6)

L'avancement de ce programme est décrit ci-après.

Par ailleurs, le programme initial supposait des investissements pour expérimentation en cellule chaude de vannes prototypes, en atmosphère tritiée pour lesquels un dossier avait été préparé. Au cours de l'étude des problèmes budgétaires (CEA et CEE) et des problèmes techniques (adoption de vannes "tout métal") ont conduit à geler toute réalisation de pilote.

#### VANNES d'ISOLEMENT (Circuit du Tore)

Les premières "Spécifications Techniques" avaient pour Objet des vannes polyvalentes qui auraient pu indifféremment équiper des circuits de vide à base de cryopompes ou de pompes turbo moléculaires. L'addition des exigences requises par les deux systèmes aboutissaient à une extrème sévérité de ces spécifications techniques.

Les consultations ont été limitées, en priorité, à des vannes d'isolement destinées à des circuits équipés de pompes turbo moléculaires pour lesquelles ont été rédigées les spécifications techniques (7ème version), les spécifications d'étude de faisabilité et sa "justification". Des appels d'offres ont été lancés auprès des industriels communautaires ; sept réponses positives ont été analysées pour établissement d'une liste réduite : AMRI (F), FIAT-TTG(I), LEYBOLD (RFA), NEYRPIC (F), NTG, RAPPOLD (RFA) et VAT (CH).

Après dépouillement des appels d'offres un rapport a été rédigé par le CEA, base de discussion en GT6 avec KFK et l'équipe du projet NET. Un rapport définitif, proposant l'attribution d'un contrat de faisabilité à la Sté Suisse VAT, a été soumis au "AD HOC GROUP ON T6" de Bruxelles et transmis, avec avis favorable, pour décision, au FTSC de BRUXELLES (séance du 1.10.1986) qui l'a entériné. Ce contrat a été signé en Février 1987.

AHG "Ad Hoc Group on T6 = CEE (Fusion Directory Bruxelles), KFK, NET, CEA.

#### 2. POMPES TURBOMOLECULAIRES

La procédure complète d'attribution d'un contrat industriel de faisabilité a été effectuée : élaboration de spécifications techniques du composant (7ème version) et d'étude de faisabilité avec sa "justification", dépouillement des appels d'offres, lancement et suivi d'une commande.

Pour les quatre industriels en compétition (ALCATEL (F), EDWAKDS (GB), LEYBOLD-HERAEUS et PFEIFFER (RFA), la Société allemande PFEIFFER collaborant avec la Société Française S2M, a été choisie, après mise au point d'un programme d'essais détaillé.

Dans le cadre du contrat qui lui a été attribué par la Communauté Européenne, PFEIFFER a effectué des mesures de débit et de taux de compression à l'hélium sur le plus gros modèle de ses pompes turbomoléculaires de série (TPH 5000) équipé de différents types de roues. Ces mesures permettront une meilleure définition de la forme des roues de la pompe turbomoléculaire, à l'étude, de 50 000 l.s $^{-1}$ ; la pompe ainsi définie possèdera 17 étages pour quatre formes différentes de roues. Ses principales caractéristiques, obtenues par calcul sont les suivantes :

- débit volumétrique hélium 50 57 000 l.s<sup>-1</sup>
- taux de compression hydrogène 1,1.107 environ
- moment d'inertie du rotor 254 kg.m<sup>2</sup>
- contrainte maximale dans les disques 180 N/mm.

Un montage a été réalisé dans la pompe TPH 5000 destiné à mesurer les efforts sur le rotor générés lors d'entrées rapides de gaz en basse pression; un rapport confidentiel d'essais, couvrant le premier semestre, a été édité en janvier 1987.

#### 3. POMPAGE CRYOGENIQUE

L'équipe du NET a lancé un appel d'offres pour une étude d'ingénièrie des circuits de vide du Réacteur de Fusion en projet dont l'objet principal est comparaison technico-économique de la solution du pompage à base de pompes cryogéniques à celle du pompage par pompes turbomoléculaires. Elle permettra une meilleure définition des circuits de vide, indispensable à la fixation des spécifications de leurs composants. Simultanément, une étude bibliographique sur "l'état de l'art" du pompage cryogénique de l'hélium (pompage par cryocondensation, cryoabsorption et par jets de gaz condensés) a été effectuée par le Groupe de Travail KFK/CEA.

#### ACTIVITES 1987

L'activité 76 prévue pour la période 1983/1986 a été prolongée en 1987.

Les activités du Groupe de Travail seront consacrées essentiellement :

- a) au lancement de l'étude des composants suivants :
  - pompes primaires (à monter en série avec les pompes turbomoléculaires)
  - vannes à fermeture rapide (isolement des pompes turbo-moléculaires)
- b) au suivi des deux contrats de faisabilité déjà attribués à des industriels européens :
  - PFEIFFER (RFA) : pompe turbo-moléculaire
  - VAT (CH) : vanne de grand diamètre
- c) au suivi des contrats concernant les circuits de vide du réacteur de fusion NET (projet européen) : comparaison du pompage cryogénique et du pompage turbomoléculaire (PFEIFFER) et études particulières de cryogénie (l'AIR LIQUIDE).

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- 2 Comptes rendus des 5 réunions du Ad Hoc Group on T6" CEA/KFK
- 3 Comptes rendus de réunions CEA/NET/KFK

PJ:

- 1 Prévisions financières initiales de l'action
- 2 Planning initial de l'action