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Some Considerations on Tritium Control in a Helium Cooled
Ceramic Blanket for the NET Reactor

Abstract

Since 1983 the Karlsruhe Nuclear Research Center is investigating the technological problems of helium cooled ceramic blankets for fusion reactors. Various solutions have been so far investigated: poloidal, toroidal or lobular arrangement of the pressure tubes containing the breeder material and the cooling helium; lead multiplier integral with the first wall or beryllium in the ceramic breeder region.

For all these designs crucial is the requirement to maintain the tritium losses from the plant equal or below 10 curie/d. This requires a helium purge flow completely separated from the main helium coolant flow. If the helium pressure in the purge system is only slightly lower than the pressure in the main coolant circuit (80 bar for the poloidal arrangement and 50 bar for the toroidal or lobular arrangement of the pressure tubes) tritium containment requires an oxidizing atmosphere in the purge flow region. With a helium purification plant of reasonable size the calculated equilibrium partial pressure of T_2O in the helium purge flow is 19 or 12 pascal for the case of 80 or 50 bar helium pressure, respectively. In case of a purge system at 1 bar no oxidizing atmosphere in the purge flow is required, provided that the primary circuit has an oxidizing helium atmosphere. This system however is based on the concept of the cladding collapsed on the ceramic pellets and it is questionable whether the pellets can support the cladding after irradiation, due to crackings caused by the thermal stresses.

The assessment of the consequences of having a T_2O partial pressure of 19 pascal in direct contact with the ceramic breeder material has been performed for Li_2O . To avoid the formation of a stable $LiOT$ phase, it is necessary to operate the Li_2O at temperatures $\gg 400^\circ C$ for $p_{T_2O} = 19$ pascal. For a NET size blanket the

Li_2O inventory is about 40 tonnes. The relative tritium inventory due to tritium diffusion is 285 grams. The tritium dissolved in the Li_2O is 21 or 16 grams for $p_{\text{T}_2\text{O}} = 19$ or 12 pascal, respectively. These values appear to be quite acceptable. The experiments of Guggi et al indicate that a certain T_2O partial pressure in helium could increase the amount of T_2O adsorbed on the surface of the Li_2O particles and thus the tritium inventory. No sufficient data is available at present to assess the amount of T_2O adsorbed.

T_2O increases the LiOT vapor pressure: for instance for 850°C and $p_{\text{T}_2\text{O}} = 19$ pascal the LiOT vapor pressure is 1 pascal and this could produce a considerable LiOT transport with possible occlusion of the channels for the helium purge flow. It is expected that this problem is much alleviated by the use of other, less hygroscopic ceramic materials such as silicates or aluminates.

Einige Betrachtungen über die Tritiumkontrolle in einem heliumgekühlten, keramischen Blanket für den NET-Reaktor

Zusammenfassung

Seit 1983 beschäftigt man sich im Kernforschungszentrum Karlsruhe mit den technologischen Problemen des heliumgekühlten, keramischen Blankets eines Fusionsreaktors.

Verschiedene Konzeptlösungen sind dazu bereits untersucht: poloidale, toroidale oder lobulare Anordnung der Druckrohre mit dem Brutstoff und Heliumkühlung; Neutronenmultiplikatoren als Blei in Verbindung mit der ersten Wand oder als Beryllium in dem keramischen Brutstoffbereich.

Unabdingbar für alle diese Konzepte ist die Forderung, den Tritiumverlust der Anlage auf 10 Curie/d oder darunter zu halten. Das bedingt einen Heliumspülgasstrom, der vom Hauptkühlkreislauf vollständig getrennt ist. Wenn der Heliumdruck im Spülsystem nur wenig geringer ist als der Druck im Hauptkühlkreislauf (80 bar bei der poloidalen Anordnung und 50 bar bei der toroidalen bzw. lobularen Anordnung der Druckrohre), dann ist es erforderlich, daß sich das Tritium im Spülgasbereich in einer oxidierenden Atmosphäre befindet. Setzt man eine Heliumreinigungsanlage in einer vernünftigen Größe voraus, dann läßt sich zeigen, daß der T_2O -Partialdruck im Spülgas von 80 bar Heliumdruck 19 Pascal bzw. bei 50 bar Heliumdruck 12 Pascal betragen wird. Im Falle des Spülgassystems bei 1 bar ist keine oxidierende Atmosphäre notwendig, vorausgesetzt, daß der primäre Kreislauf eine oxidierende Heliumatmosphäre besitzt. Dieses System basiert jedoch auf dem Konzept, bei dem die Hülle auf das Keramikmaterial kollabiert und es ist fraglich, ob die Pellets nach der Bestrahlung, bei der Risse durch thermische Spannungen entstehen, das Hüllmaterial stützen können.

Es wurden auch die Folgen eines direkten Kontaktes des keramischen Li_2O -Brutstoffs mit T_2O bei einem Partialdruck von 19 Pascal untersucht. Damit vermieden wird, daß sich eine stabile $LiOT$ -Phase bildet, ist es bei Li_2O und $p_{T_2O} = 19$ Pascal notwendig oberhalb $400^\circ C$ zu arbeiten. Für ein Blanket der NET-Größe bedarf man ein

Li₂O-Inventar von etwa 40 Tonnen. Das Tritiuminventar dieses Blankets bedingt durch die T-Diffusion beträgt 285 g. Das gelöste Tritiuminventar in diesem Li₂O-Inventar beträgt bei $p_{T_2O} = 19$ Pa gleich 21 g und bei $p_{T_2O} = 12$ Pa gleich 16 g. Diese Werte erscheinen ganz annehmbar. Die Untersuchungen von Guggi u.a. deuten an, daß sich bei einem bestimmten T₂O-Partialdruck in Helium das Tritiuminventar durch T₂O-Adsorption an der Oberfläche der Li₂O-Partikel erhöhen könnte. Zur Zeit sind noch nicht genügend Daten dazu vorhanden, um die adsorbierte T₂O-Menge abschätzen zu können.

T₂O erhöht den LiOT-Dampfdruck; z.B. ist der LiOT-Dampfdruck bei $p_{T_2O} = 19$ Pascal gleich 1 Pascal. Das könnte zu einem beträchtlichen LiOT-Transport und damit zur Verstopfung der Kanäle des Heliumspülgases führen. Es ist zu erwarten, daß dieses Problem mit weniger hygroskopischem Keramikmaterial, wie z.B. mit Silikaten oder Aluminaten, vermindert wird.

1. Introduction

A conceptual design study has shown that ceramic lithium containing material with helium cooling could be a very attractive solution as a tritium breeding blanket of a fusion reactor /1/. These first investigations have been performed with the boundary conditions, such as blanket dimensions, temperatures and so on, suggested by the INTOR study /2/. This blanket design was based on a lead multiplier integral with the stainless steel first wall and on a breeder material in form of Li_2SiO_3 spheres contained in two meters long pressure tubes running in toroidal direction. This toroidal solution is being pursued further, in more detail, in collaboration with the Hauptabteilung Ingenieurtechnik of the Karlsruhe Nuclear Research Center. After a suggestion of the NET team /3/ we are now investigating a similar solution with pressure tubes running in a poloidal direction. First thermohydraulic calculations have been performed /4/. After the publication of the American "Blanket Comparison and Selection Study" /5/ we are investigating the so-called "lobular" arrangement of the blanket, which has been suggested by General Atomic /6/. These design studies are performed in collaboration with Interatom. To avoid the temperature limitations due to lead, we have lately started to study the use of beryllium as multiplier.

In the arrangement described in Ref. /1/ and in some of the other considered solutions, the helium coolant flow was used as a purge flow for the tritium as well, the tritium containment being based on the assumption that an oxidizing helium atmosphere was capable of transforming all the present tritium in tritiated water, thus avoiding the permeation of molecular tritium through metal walls at high temperatures. This problem is dealt in more detail in the present report.

The considerations illustrated in the present report are based on the poloidal solution with a separate lead multiplier mentioned above /4/. The main data of this blanket design are given in Table I. However these considerations are of rather general nature and are only slightly affected by the particular considered blanket design.

2. Tritium Containmentment within the Primary Helium Circuit

2.1 Solution with No-separate Helium Purge Flow

Assuming as a in Ref. /1/ a total volume of the helium primary circuit equal to 3200 m^3 (the same as that of the German helium cooled prototype reactor THTR /7/) and the helium pressures and temperatures given in Table I, we obtain a total helium inventory in the primary helium circuit of 24500 kg of helium. The total mass flow of the coolant helium for the data of Table I is 400 kg/sec. It is estimated that 65% of this helium flow is required by the first wall-multiplier and breeder sections of the blanket, the rest being used to cool the shields, divertor and other structures in the blanket region. The helium coolant flow in the blanket region is thus 260 kg/sec. Generally in helium cooled fission reactors a small amount of the helium flow (slipstream fraction) is diverted to a helium purification plant, where the various impurities contained are separated from the helium, and then reintroduced in the main helium circuit. We may assume for the present considerations that the slipstream fraction is 0.1% of main helium flow in the in first wall-multiplier and breeder sections of the blanket, i.e. it is equal to 0.26 kg/sec. This value is similar to the flow which was foreseen in the Unit 1 of the purification plant of the Gas-Cooled Fast Breeder Reactor Demonstration Plant of 300 MWe, designed by General Atomic /8/, i.e. 0.2 kg/sec, Unit 2 being designed for about the half of this helium flow. These plants are similar, in particular Unit 1, to the Helium Purification System operating in the High Temperature Helium Cooled prototype of Fort St. Vrain. Under these conditions the average residence of the helium in the primary circuit, therefore of the tritium contained in it, is given by:

$$t_R = \frac{24500}{0.26} \approx 10^5 \text{ sec} \quad (1)$$

We may assume that the tritium production in this reactor is the same as that calculated for the toroidal solution described in Ref./1/, i.e. 88 grams of tritium per full operating day. This is equivalent to 10^{-3} grams of tritium per second, i.e. the equilibrium tritium inventory in the helium primary circuit is in this case equal to:

$$I_{\text{THe}} = 10^5 \times 10^{-3} = 100 \text{ g} = 10^6 \text{ curie} \quad (2)$$

Tritium may emerge from irradiated ceramic material as molecular T_2 or as tritiated water (T_2O). The best known ceramic material is Li_2O . Japanese studies indicate that under vacuum 95 to 98% of the tritium is produced in form of T_2O , the rest being T_2 /9/. Other experiments performed at Oak Ridge with γ - $LiAlO_2$ in a helium purge flow containing 0.1% hydrogen (reducing atmosphere) show that tritium is recovered in helium in the HT form, the ratio HT to HTO being about 1000 to 1 /5/. To estimate the tritium losses we will thus perform calculations assuming that the tritium is either available in T_2 form or in T_2O . The first situation is probably achievable with a reducing atmosphere (H_2 in helium) and the second with a slightly oxidizing atmosphere (O_2 in helium). The chemical form under which tritium is available in the helium primary circuit is very important because tritium, like hydrogen, has a high permeability through metal walls at high temperatures, while T_2O , like H_2O , has a much lower permeability.

Most of the surface available for tritium leakage is given by the surface of the heat exchangers between primary helium circuit and secondary water-steam circuit. Of course it is not yet decided to produce steam and electrical energy with the NET reactor, in any case however the blanket should be prototypical that is capable to operate in a situation where electrical energy is produced. For our estimate we have assumed that the total surface

of the helium primary circuit of NET (620 MW of power) is the same as the total surface of the Gas-Cooled Fast Breeder Reactor Demonstration Plant of 300 MWe, which has similar if slightly higher power (830 MWth), a similar helium pressure (88 bar) and a similar, if slightly higher helium temperature range (320 - 550°C). The relevant data for this reactor are:

- heat exchangers total surface = 10^4 m^2
- heat exchangers material = incoloy 800
- heat exchanger wall average thickness = 1.86 mm /8/.

2.1.1 Tritium Losses by Permeation with a Reducing Atmosphere

In a reducing atmosphere the 100 g of tritium are available as T_2 100 grams of tritium are equivalent to $\frac{100}{6} = 16.7$ Mol of T_2 . The number of moles of helium in the primary circuit is given by:

$$n_{\text{He}} = \frac{3200 \times 80}{22.4 \times 10^{-3}} \frac{293}{\frac{80+380}{2} + 273} = 6.66 \times 10^6 \quad (3)$$

and the T_2 partial pressure is

$$p_{T_2} = \frac{16.7}{6.66 \times 10^6} 80 = 2 \times 10^{-4} \text{ bar} = 20 \text{ pascal} = 0.152 \text{ torr} \quad (4)$$

The permeation of tritium through incoloy 800 has been measured by Bell and Redman /10/, who have obtained the following equation valid in the range 500 to 1000 K:

$$V_{T_2} \left[\frac{\text{cm}^3 (\text{STP})}{\text{min}} \right] = 0.566 e^{-\frac{8141}{T [\text{K}]}} \times \frac{A [\text{cm}^2]}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.5} \quad (5)$$

In our case the helium temperature varies between 80°C and 380°C, and we may assume with good approximation that the heat exchanger surface temperature varies linearly between $T_1=80^\circ\text{C}=353 \text{ K}$ and $T_2=380^\circ\text{C}=653 \text{ K}$. I.e.:

$$T = T_1 + \frac{T_2 - T_1}{A_0} A \quad (6)$$

with $A_0 = 10^4 \text{ m}^2 = 10^8 \text{ cm}^2$.

and
$$dA = \frac{A_0}{T_2 - T_1} dT \quad (7)$$

Thus the resulting tritium leakage flow may be calculated as:

$$\begin{aligned} V_{T_2} \left[\frac{\text{cm}^3 (\text{STP})}{\text{min}} \right] &= \frac{0.566}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.5} \int_{T_1}^{T_2} \left(e^{-\frac{8141}{T}} \frac{A_0}{T_2 - T_1} \right) dT = \\ &= \frac{0.566}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.5} \frac{A_0}{T_2 - T_1} \left[\frac{e^{-cx}}{x} + c \text{ Ei}(-cx) \right]_{x_1}^{x_2} \quad (8) \end{aligned}$$

with $c = 8141$

$$x = \frac{1}{T}, \quad x_1 = \frac{1}{353}, \quad x_2 = \frac{1}{653}$$

Ei(-cx) is a function of -cx which is tabulated in Ref. /11/

With the values chosen or calculated in the present and previous section i.e. $p_{T_2} = 0.152$ torr, $A_0 = 10^8$ cm², $s = 1.86$ mm, $T_1 = 353$ K, $T_2 = 653$ K we finally obtain.

$$V_{T_2} = 28.1 \text{ cm}^3(\text{STP})/\text{min} = 28.1 \times \frac{6}{22.4 \times 10^3} 10^4 \times 60 \times 24 = 1.08 \times 10^5 \text{ curie/d} \quad (9)$$

This figure is much too high, if we consider that we should keep the amount of tritium loss from the plant lower or equal to 10 curie/d (see for instance Ref. /12/).

The relationship of Bell and Redman (Eq.(5)) was obtained for a blank metallic surface. It is known that an oxide film on the surface may reduce quite considerably the tritium permeation. In presence of a reducing atmosphere in the primary helium circuit it is quite unlikely that an oxide film is formed on the helium side of the heat exchanger surface, however, due to the oxidizing effect of the steam, an oxide layer may be formed on the steam side of this surface. To assess the effect of this oxide film barrier we make use of the experimental information obtained by Strehlow and Savage /13/. These authors show that the permeation of deuterium through incoloy 800 which has been exposed to H₂O at 538°C and 240 bar (typical conditions for superheated steam for a modern turbine) for 1000 hours is given at 531°C (=804 K) by the following equation:

$$V_{D_2} \left[\frac{\text{cm}^3(\text{STP})}{\text{h}} \right] = 6.3 \times 10^{-4} \times \frac{A \text{ [cm}^2\text{]}}{s \text{ [mm]}} \left(p_{D_2} \text{ [torr]} \right)^{0.51} \quad (10)$$

Furthermore Strehlow and Savage state that their data for most of the alloys investigated give heats of activation for permeation of 17 to 18 Kcal/mole. If we assume an heat of activation of 17.5 Kcal/mole = 73250 J/mole, correct for the difference in diffusivity between D₂ and T₂ with the factor $\sqrt{2/3}$ /14/, and remember that the gas constant R is 8.314 J/mole, we may write:

$$\begin{aligned}
 V_{T_2} \left[\frac{\text{cm}^3 (\text{STP})}{\text{h}} \right] &= \sqrt{\frac{2}{3}} \times 6.3 \times 10^{-4} \frac{A [\text{cm}^2]}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.51} e^{-\frac{73250}{8.314} \left(\frac{1}{T} - \frac{1}{804} \right)} \\
 &= 29.56 \frac{A [\text{cm}^2]}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.51} e^{-\frac{8811}{T}}
 \end{aligned} \tag{11}$$

For a linear distribution of temperature on the heat exchanger surface, Eq. (11) may be integrated in same way as Eq. (5). We obtain an equation similar to Equation (8). V_{T_2} may then be calculated for $p_{T_2} = 0.152$ torr, $A_0 = 10^8 \text{ cm}^2$, $s = 1.86$ mm, $T_1 = 353$ K, $T_2 = 653$ K. We obtain:

$$V_{T_2} = 107 \frac{\text{cm}^3 (\text{STP})}{\text{h}} = 107 \times \frac{6 \times 10^4}{22.4 \times 10^3} \times 24 = 0.69 \times 10^4 \text{ curie/d} \tag{12}$$

A value which is a factor 16 lower than the previous, but still much higher than the required value of 10 curie/d.

2.1.2 Tritium Losses by Permeation and Helium Leakage with an Oxidizing Atmosphere

A very effective way to reduce the tritium leakage is the use of an oxidizing atmosphere in the helium primary circuit by maintaining for instance a certain oxygen partial pressure there. This suggestion has been already put forward by General Atomic /5/. In equilibrium conditions the tritium partial pressure may be calculated by the following expression:

$$p_{T_2} [\text{bar}] = \frac{p_{T_2O} [\text{bar}]}{K \sqrt{p_{O_2} [\text{bar}]}} \tag{13}$$

where K is the reaction constant, function of the gas mixture temperature. The kinetic of the reaction in the temperature range we are considering may be very slow indeed, so that equilibrium conditions may be not representative of the real case. To reach equilibrium we may need a catalyzer or a solid oxidizing agent such as CuO which should be placed directly in the blanket region. This problem should be investigated further. For the moment, we assume that the reaction of Equation (13) is governed by the highest temperature in the helium circuit, i.e. 380°C. This is because at the highest temperature the reaction is the fastest and because this is the temperature at the point where helium is leaving the blanket, furthermore at this temperature the resulting tritium partial pressure is the highest, i.e. our calculation is pessimistic. For T=380°C the reaction constant K is equal to 1.78×10^{17} bar^{1/2} [15]. We could then provide an amount of oxygen capable of oxidizing the tritium which is emerging from the breeding material in T₂ form and maintain a certain surplus of free oxygen, for instance an oxygen partial pressure of 10 pascal. For these conditions the resulting equilibrium partial pressure of T₂ at 380°C is:

$$\begin{aligned}
 P_{T_2} &= \frac{P_{T_2O}}{K \sqrt{P_{O_2}}} = \frac{2 \times 10^{-4}}{1.78 \times 10^{17} \sqrt{10^{-4}}} = 1.12 \times 10^{-19} \text{ bar} = \\
 &= 0.85 \times 10^{-16} \text{ torr}
 \end{aligned}
 \tag{14}$$

This is of course an extremely low pressure, and it is quite clear that in this case the tritium permeation losses from the primary circuit must be very small. An estimate of these losses can be obtained using, again, the data of Strehlow and Savage /13/, because it is quite likely that, with an oxidizing atmosphere in the helium primary circuit, an oxide film is present on the heat exchanger surface. The resulting tritium loss is of the order of 10^{-14} curie/d. It should be stressed that this is only

a rough estimate, because the relationships of Ref. /13/ are valid for a pressure range down to 10^{-3} torr and we have applied them to a pressure of 10^{-16} torr. The calculated tritium losses however are so small that we may conclude that they are in this case negligible. Even the use of the equation of Bell and Redmann (blank metal surface) would produce a tritium loss by permeation of only 10^{-3} curie/d.

At this point one should notice that in our considerations we have neglected the presence of other impurities, besides T_2O , T_2 , O_2 , in helium. Other impurities, such as H_2 , H_2O , N_2 , CO , CO_2 , CH_4 , could in principle be present in the helium primary circuit. Generally these gases are adsorbed on the surfaces of the circuit or in the pores, especially of ceramic materials. These gases, at high temperature and in presence of a very dry helium atmosphere, tend to be released to the gas phase. By the operation of helium cooled fission reactors these impurities are generally separated from the coolant helium by the helium purification plant, during the plant commissioning period i.e. before the reactor is brought up to full power. The amount of impurities may then reach the value of a few vpm for reactor full power operation: for Fort St. Vrain a total amount of impurities of 17 vpm is quoted /16/, while Ref. /17/ gives for the Dragon reactor a total amount of impurities less than 0.6 vpm. It is conceivable that before starting to produce tritium in the blanket one should circulate hot dry helium in the primary circuit, separating continuously the impurities contained in helium by means of the helium purification plant. To heat up the helium is not necessary to operate the fusion reactor, the required heat being produced by the helium blowers.

The oxidizing atmosphere in helium appears then to keep the tritium losses within the limit of 10 curie/d. Another problem remains however, and this the problem of the helium losses by leakage from the primary circuit. The helium primary circuit of a reactor of about 600 MWth power is very big and complex: as we

have already mentioned the surface of the heat exchangers is of the order of ten thousand square meters. To avoid completely a helium leakage from such a system is in our opinion an impossible engineering requirement. The gas losses from big gas-cooled fission reactors are of the order of a few percent (up to 10%) of the helium inventory per day. The Dragon reactor is a noticeable exception to this rule. This reactor was conceived when the coated particles were not yet invented. It was therefore believed that the primary helium circuit radiation activity would be much higher than the one which was really achieved by the use of the fuel containing coated particles. Great effort therefore was concentrated on the problem of reducing helium losses, and therefore radiation losses, from the primary circuit. The objective was to achieve a helium leak rate of 0.1% per day. Operation of the Dragon reactor showed that the achieved leak rate was better than the objective: namely it lay between 0.025 and 0.05%/d /17/. For the present design of helium cooled blanket we shall therefore assume these values, having in mind that this requires special precautions and a high engineering standard. Although very small these helium losses mean an excessive tritium loss, indeed the tritium inventory in helium is 10^6 curie (see Eq. (2)) and these losses are equivalent to 250 - 500 curie/d tritium leakage, which are still considerably higher than the required level of 10 curie/d.

It seems that the problem of the tritium losses cannot be solved even by almost complete oxidation of the tritium produced. A possible alternative would be of course to reduce the tritium inventory contained in helium. This however would require an increase of the slipstream fraction in the purification plant of a factor of 25 to 50. This of course would mean a helium purification plant 25 to 50 times bigger than that of the Fort St. Vrain reactor, i.e. a very high cost. Furthermore in the purification plant helium is cooled, expanded and then heated and compressed, and such a large purification plant would require a great amount of energy with considerable decrease of the net efficiency of the plant.

A more reasonable alternative seems to be to separate the main helium coolant flow from the helium purge flow, whose main function is to carry away the tritium produced in the ceramic breeding material. This type of solution is the one adopted in the American designs /5/. There are of course other reasons which favour the use of a separate purge system. For instance, during an accident or a maloperation of the blowers, a large amount of water or steam could get into the helium primary circuit. This could greatly dilute the tritium and make the tritium separation much more difficult.

2.2 Solution with a Separate Helium Purge Flow

The total flow of helium in a separate helium purge system goes to the helium purification plant. Let us assume for the present considerations a total helium mass flow again of 0.26 kg/sec for the same reasons mentioned in Section 2.1. For a given helium mass flow it is possible to calculate the average velocity of the helium purge flow once helium pressure, temperature and flow cross section are known. We shall perform the calculation for the poloidal solution of Ref. /4/. In this case the blanket breeder material is made up of equal size pebbles with a filling factor of about 63%. The average helium pressure is 80 bar, the minimum and maximum helium temperatures are 235°C and 750°C respectively (Table I). The resulting helium velocity in the particle bed is 1.75 cm/sec. The total length of the bed region in the blanket is 600cm, thus the helium residence time in the particle bed is $600/1.75 = 340$ sec. This may be assumed to be the total residence time of the helium in the helium purge system because obviously the helium velocity in the rest of the circuit is much higher. With this residence time and a tritium production of 10^{-3} grams of tritium per second we obtain a tritium inventory in helium of 0.34 g, i.e. 0.0564 moles of T_2 . Knowing the total helium volume in the breeder region ($V = 1.77 \times 10^4$ liter /4/) and the helium average temperature there, we finally obtain the partial pressure of T_2 (or T_2O) in the breeder region:

$$p_{T_2} \text{ (or } p_{T_2O}) = 0.0564 \frac{22.4}{1.77 \times 10^4} \frac{765.5}{293} = 1.9 \times 10^{-4} \text{ bar} = 19 \text{ pascal} = 0.144 \text{ torr} \quad (15)$$

This partial pressure may be calculated for other helium pressures in the following way. Consider two different blankets at different helium pressures and of different length. For a given helium mass flow, a given tritium production, a total helium volume in the breeder region and helium average temperature, the relationship between the helium average velocities is given by:

$$v_1 = v_2 \frac{p_2}{p_1} \frac{A_2}{A_1} = v_2 \frac{p_2}{p_1} \frac{V_2}{V_1} \frac{L_1}{L_2} = v_2 \frac{p_2}{p_1} \frac{L_1}{L_2} \quad (16)$$

and the two helium blanket residence times are related by the equation:

$$\frac{t_1}{t_2} = \frac{L_1}{v_1} \cdot \frac{v_2}{L_2} = \frac{L_1}{L_2} \frac{p_1}{p_2} \frac{L_2}{L_1} = \frac{p_1}{p_2} \quad (17)$$

Thus the ratio of the two tritium partial pressures and inventories is:

$$\frac{p_{T_2 1}}{p_{T_2 2}} = \frac{I_1}{I_2} = \frac{t_1}{t_2} = \frac{p_1}{p_2} \quad (18)$$

where: v = average helium velocity in breeder region
p = helium pressure in purge flow system
A = cross section area for helium purge flow in breeder region
V = total helium volume in breeder region
L = length of breeder region
t = residence time of helium (and of tritium) in breeder region
I = tritium inventory in the helium of the breeder region

p_{T_2} = tritium partial pressure in helium in the breeder region

Using Eq.(18) and knowing that $p_{T_2}=19$ pascal for $p=80$ bar, we obtain a tritium partial pressure of 12 pascal for an helium pressure of 50 bar (toroidal and lobular pressure tube arrangement) and a tritium partial pressure of 0.24 pascal for a helium pressure of 1 bar in the purge system.

2.2.1 Tritium Losses from the High Pressure (80 bar) Helium Purge System to the Primary Helium System

We shall perform the calculation for the purge flow system and primary helium system at 80 bar only, the results with 50 bar being rather similar. The surface between these two systems has been estimated at $8 \times 10^7 \text{ cm}^2$ with average wall thickness 0.5 mm /4/. We must distinguish the case of reducing or oxidizing atmosphere in the helium purge flow.

In the case of reducing atmosphere in the purge flow we may apply the following equation obtained by Bell and Redman for stainless steel 316:

$$V_{T_2} \left[\frac{\text{cm}^3 (\text{STP})}{\text{min}} \right] = 0.533 e^{-\frac{8212}{T}} \frac{A [\text{cm}^2]}{s [\text{mm}]} \left(p_{T_2} [\text{torr}] \right)^{0.5} \quad (19)$$

For $A=8 \times 10^7 \text{ cm}^2$, $s=0.5 \text{ mm}$, $p_{T_2}=19 \text{ pascal} = 0.144 \text{ torr}$, $T_1=180^\circ\text{C}=453 \text{ K}$, $T_2=380^\circ\text{C}=653 \text{ K}$ (see Table I), after integration as in Eq.(8), we obtain the tritium losses from the purge system to the primary helium system: $V_{T_2}=54600 \text{ curie/d}$. If the helium atmosphere in the primary circuit is reducing, then we may calculate the tritium losses to the steam circuit with Eq.(8) and obtain a tritium leak rate of 21300 curie/d. If the helium atmosphere in

the primary circuit is oxidizing the tritium losses by permeation are negligible, however the tritium losses due helium leakage (0.025-0.05%/d of the tritium inventory in the helium primary circuit = $\frac{54600 \times 10^5}{3600 \times 24} = 0.632 \times 10^5$ curie) amount to 16 to 32 curie/d.

In the case of an oxidizing atmosphere in the purge flow the tritium losses from the purge system to the primary helium system by permeation are negligible. The losses due to helium leakage from the purge flow to the primary helium circuit (0.025-0.05%/d of the tritium inventory in the purge system = 0.34×10^4 curie) amount to 0.9 ÷ 1.8 curie/d.

The main results of the calculations of this section are given in Table II. From the table it is clear that the only possibility of having a tritium loss from the plant less than 10 curie/d is to have an oxidizing atmosphere in the helium purge flow system.

2.2.2 Tritium Losses from the Low Pressure (1 bar) Helium Purge System to the Primary Helium System

The calculations are similar to those of the previous section, with the difference that the tritium partial pressure in the helium purge system is 0.24 pascal and the tritium inventory in the helium purge system in the blanket region is 44 curie. The main results of the calculations are given in Table III. It may be seen from the table that, in this case, to have a tritium loss from the plant less than 10 curie/d, it is not necessary to have an oxidizing atmosphere in the helium purge flow system, provided that the primary circuit has an oxidizing helium atmosphere.

3. Consequences for the Ceramic Breeder Material of Having an Oxidizing Atmosphere in the Helium Purge Flow

The main conclusion of the previous Section 2 is that, to maintain the tritium losses below 10 curie/d, it is necessary to have a certain partial pressure of T_2O in the range of 10 to 20 pascal in the helium atmosphere surrounding the breeding ceramic material. The only possibility to avoid this is to have a helium purge flow system at 1 bar. This system however is based on the concept of the cladding collapsed on the ceramic pellets /5/ and it is questionable whether the pellets can support the cladding after irradiation due to crackings caused by the thermal stresses. Furthermore a sphere pac type of ceramic fuel is much better suited to fill up the available space in the calandria type fuel container foreseen in our design /4/. This is particularly the case, when the use of beryllium multiplier mixed with the breeder material is envisaged.

The assessment of the consequences of having a certain T_2O partial pressure (12 to 19 pascal) in direct contact with the ceramic breeder material has been performed for lithium oxide (Li_2O), because this is the only lithium containing ceramic material for which the relevant quantitative information is available. For other lithium containing ceramic material of interest (aluminates, silicates) only qualitative inferences can be made, based on the knowledge that these materials are less hygroscopic than Li_2O . The information available for Li_2O refers to the effects of H_2O . We assume here that for the phenomena which we are considering no difference exists between H_2O and T_2O , because these refer mainly to chemical behaviour.

3.1 Formation of a Stable LiOT Phase

The formation of a stable LiOT phase should be avoided because this would imply an excessive tritium inventory in the ceramic. For instance for an initial Li_2O inventory of 40 tonnes /4/, a complete transformation to LiOT would be in principle possible and this would mean a tritium inventory of 8 tonnes, which is unacceptable.

The system H_2O , Li_2O , LiOH has been studied by Tetenbaum and Johnson /18/. Fig.1 shows the results of their experimental investigation. A H_2O partial pressure of 19 pascal corresponds to a temperature of 387.5°C on the line separating the stable LiOH phase from the Li_2O phase. This means that, for this H_2O partial pressure, at temperatures lower than 387.5°C the LiOH phase is the stable one and for temperatures higher than 387.5°C LiOH is not stable and can be present in equilibrium only in solution with Li_2O . This of course implies that some tritium would be dissolved in the lithium oxide. We will deal with this question in the next section. To have a sufficient margin we would therefore suggest a minimum Li_2O temperature in the blanket of 400°C for a T_2O partial pressure of 19 pascal.

Fig. 1 shows that for $p_{\text{H}_2\text{O}} = 12$ pascal the phase separation temperature is 375°C . However for $p_{\text{H}_2\text{O}} = 0.24$ pascal this temperature is only 282.5°C .

It is conceivable that with lithium aluminates and silicates these separation temperatures should be lower. So far no information is available to us for these materials. Our calculations show that, with a beryllium multiplier, it is possible to keep the ceramic material in the temperature range $400 \div 850^\circ\text{C}$ /4/.

3.2 Solubility of LiOT in Li₂O

Even for temperatures above the phase separation line LiOT could be present in the Li₂O breeder material, not as a separate phase but in solution. This of course would imply a certain amount of tritium dissolved in the breeder material. This amount may be estimated assuming that the solubility of LiOT in Li₂O is the same as the solubility of LiOH in Li₂O. The solubility of LiOH in Li₂O has been measured in Argonne for various temperatures and H₂O partial pressures in helium /19/. These experimental data have been correlated by the following analytical expression:

$$\log(x_{\text{LiOH}}) = (0.427 + 1.7 \times 10^{-4}T) \log(p_{\text{H}_2\text{O}}) - 17.667 + 2.502 \times 10^{-2}T - 9.62 \times 10^{-6}T^2 \quad (20) \quad /5/$$

where x_{LiOH} is the mole fraction of LiOH dissolved in Li₂O, $p_{\text{H}_2\text{O}}$ is the partial pressure of H₂O in helium in atmospheres, and T is the temperature in Kelvin. For the poloidal type of blanket with beryllium multiplier which we are now investigating /4/, the temperature of the ceramic breeder material lies between 400°C and 850°C. We may integrate Equation (20) with the assumption that the temperature distribution in the ceramic breeder material is linear with temperature. This assumption is pessimistic i.e. produces too high dissolved LiOT quantities, because in our design the temperature distribution around the coolant tubes in the calandria is quadratic and greater portions of breeder material will be at the higher temperatures than it is predicted by a linear temperature distribution. Equation (20) is integrated with a procedure analogous to that used to obtain Eq.(8). For a total mass of Li₂O in the blanket of 40 tonnes /4/ the total amount of tritium dissolved is:

$$\begin{aligned} 21 \text{ grams for } p_{\text{T}_2\text{O}} &= 19 \text{ pascal} \\ 16 \text{ grams for } p_{\text{T}_2\text{O}} &= 12 \text{ pascal} \\ 1.7 \text{ grams for } p_{\text{T}_2\text{O}} &= 0.24 \text{ pascal} \end{aligned} \quad (21)$$

These are very small tritium inventories and perfectly acceptable. We may compare these to the tritium inventories due to tritium diffusion in the particles. Assuming that 90% of the Li_2O is formed by pebbles of 2 mm in diameter and 10% of 0.5 mm diameter pebbles, we have that the tritium inventory due to diffusion is given by:

$$I_D = \frac{\dot{m}}{60 D} (K_1 d_1^2 + K_2 d_2^2) \quad (22)$$

with \dot{m} = tritium production rate = 0.68×10^{-3} g/sec /4/

$$K_1 = 0.9$$

$$d_1 = 0.2 \text{ cm}$$

$$K_2 = 0.1$$

$$d_2 = 0.05 \text{ cm}$$

$$D = e^{-5.93 - \frac{81730}{8.314T}} \text{ (cm}^2\text{/sec)} \quad /20/$$

D being the diffusivity of tritium in Li_2O as a function of temperature. Also Eq. (22) may be integrated with the assumption of linear variation of temperature over the Li_2O volume with a procedure analogous to that used to obtain Eq.(8). The calculated tritium inventory due to diffusion for the temperature range $T_1=400^\circ\text{C}$, $T_2=850^\circ\text{C}$ is 285 grams. Also this is an acceptable value.

3.3 T₂O Adsorption on the Ceramic Breeder Particle Surface

The experiments of Guggi et al /20/ show that the rate of tritium release from single crystal Li_2O is affected by the presence of traces of moisture (≈ 1 vpm H_2O) in the sweep helium gas. However their experiments with dry helium show quite clearly that the rate determining step is the tritium diffusion in the particles. A plausible explanation of this difference is that some moisture remains adsorbed on the surface of the particle. The data of Ref./20/ are analysed in detail in the Appendix of this paper. The objective

of this analysis was to obtain information on the adsorption rate constant. Knowledge of this adsorption rate constant would allow the calculation of the tritium adsorbed on the particle surface in the blanket. Unfortunately the analysis of the Appendix shows that the determination of this constant is not possible from the data of Ref. /20/, because of another phenomenon which covers the adsorption effects, namely isotope exchange between the tritium adsorbed on the surface and the hydrogen contained in the moisture. To obtain information on the adsorption rate constant is probably necessary to perform tests with well defined partial pressures of T_2O in helium and on the same time trying to keep the partial pressure of H_2O as small as possible.

Another indication that surface adsorption effects may be important in determining the tritium blanket inventory in presence of not negligible T_2O partial pressures may be inferred from the fact that the diffusivity values obtained by the Jülich group for lithium oxide and two lithium aluminates /20,21,22/ are generally higher of other experimental values obtained with powders in not very well defined conditions (see for instance the different values of diffusivity collected in Ref. /23/). The samples used by the Jülich group were generally rather large , either single crystal or with a very low porosity, and the helium sweep gas was dry. It is conceivable that the apparent too low values of diffusivity were determined by experiments where the tritium diffusivity in the particle was not the only one rate determining process but a considerable surface effect was present as well.

According to H.R. Ihle these surface effects should be smaller for silicates and especially for aluminates than for lithium oxide /24/.

3.4 T₂O Effects on LiOT Vapor Pressure

The experimental investigations of Tetenbaum and Johnson /25/ show clearly that the presence of a H₂O partial pressure in the helium sweep gas increases the LiOH vapor pressure. This data are correlated by the following Equation:

$$\log(p_{\text{LiOH}} [\text{Atm}]) = - \frac{8365}{T[\text{K}]} + 0.5 \log (p_{\text{H}_2\text{O}} [\text{Atm}]) + 4.57 \quad (23)$$

We may assume that this is valid for the system LiOT/T₂O as well. Table IV shows the values of the LiOT vapor pressure calculated with Eq. (23) for the highest and lowest temperatures foreseen for the breeder material and for the three different T₂O partial pressures considered in the present study. For T=400°C the LiOT vapor pressures are very low, however for T=850°C these pressures are in the region of 1 pascal for the high pressure helium purge flow systems. This value is probably too high and may lead to excessive transport of LiOT (or LiOH) from the hotter to the cooler regions of the blanket, with the possible consequence of occluding the channels available in the sphere-pac bed for the flow of the purge helium. Also the value of 0.1 pascal relative to the purge flow system at 1 bar may be too high. It is expected that this problem is much alleviated by the use of other, less hygroscopic ceramic materials such as silicates and especially aluminates.

4. Conclusions and Recommendations

Calculations have been performed to investigate the requirement of maintaining the tritium losses from the plant equal or below 10 curie/d. To obtain this, the following conditions must be fulfilled:

1. The helium purge flow must be completely separated from the main helium coolant flow of the primary helium circuit.
2. For a high pressure purge system, tritium containment requires an oxidizing atmosphere in the purge flow region. The presence of a catalyst or an oxidizing agent should guarantee that equilibrium conditions between T_2 , O_2 and T_2O are achieved. With a helium purification plant of reasonable size (helium mass flow = 0.26 kg/sec) the equilibrium partial pressure of the T_2O in the purge system is 19 or 12 pascal for the case of 80 or 50 bar helium pressure respectively.
3. In case of a purge system at 1 bar, no oxidizing atmosphere in the purge flow is required, provided that the primary circuit has an oxidizing atmosphere. This system, however, is based on the concept of the cladding collapsed on the ceramic breeder material pellets and it is questionable whether the pellets can support the cladding after irradiation, due to crackings caused by the thermal stresses and by the thermal cycling.

The assessment of the consequences of having a certain T_2O partial pressure above the ceramic breeder material has been performed for Li_2O , because this is the only lithium containing ceramic material for which the relevant quantitative information is available. The main conclusions of this assessment are the following :

4. To avoid the formation of a stable LiOT phase, it is necessary to operate the Li_2O at temperatures $\gg 400^\circ\text{C}$ for $p_{\text{T}_2\text{O}} = 19$ pascal. Silicates and especially aluminates may have a less strict requirement.
5. For a NET size blanket with a Li_2O inventory of 40 tonnes, made up of 2 mm (90%) and 0.5 mm (10%) pebbles, the tritium inventory due to tritium diffusion in the ceramic is about 300 grams. The tritium dissolved in the Li_2O is 21 or 16 grams for $p_{\text{T}_2\text{O}} = 19$ or 12 pascal respectively. These values appear to be quite acceptable.
6. There are indications from the literature that a certain T_2O partial pressure in helium could increase the amount of T_2O adsorbed on the surface of the Li_2O particles and thus increase the tritium inventory. No sufficient data is available at present to assess the amount of the T_2O adsorbed. This problem requires further investigation.
7. T_2O increases the LiOT vapor pressure: for instance at a temperature of 850°C and a T_2O partial pressure of 19 pascal the resulting LiOT vapor pressure is one pascal. This could produce an excessive LiOT transport with possible occlusion of the channels for the helium purge flow. It is expected that this problem is much alleviated by the use of other, less hygroscopic ceramic materials such as silicates or aluminates.

Acknowledgement

The computer calculations mentioned in the Appendix have been performed by C. Ferrero.

Appendix: Mathematical treatment of the tritium inventory in a spherical particle due to diffusivity and surface adsorption. Analysis of the data of Guggi et al. /20/

Guggi et al /20/ have measured the rate of tritium release from single crystal Li_2O . They show that with a dry sweep helium gas the rate determining step is the tritium diffusion in the particles. However in presence of traces of moisture (≈ 1 vpm H_2O) in the sweep helium gas the tritium release rate does not follow the time dependence dictated by a diffusion controlled phenomenon. We believe that this discrepancy is due to the fact that some moisture (T_2O or H_2O) remains adsorbed on the surface of the particle. To try to explain this and have some information on the adsorption rate constant we analyzed the data of Ref. /20/. The fractional tritium release from the particle is defined by:

$$F(t) = 1 - \frac{\bar{c}(t) - c_f}{c_o - c_f} \quad (24)$$

where c_o = initial concentration of tritium in the particle
 c_f = final concentration
 $\bar{c}(t)$ = average concentration at the time t

It can be shown that the fractional release $F(t)$ from a spherical particle with initial uniform volume concentration and zero concentration at the surface is given by:

$$F(t) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2 \pi^2 \lambda t} \quad (25) /26/$$

where $\lambda = D/R^2$
 D = tritium diffusivity in the particle, supposed uniform
 R = particle radius
 t = time

This rather complex function may be approximated by:

$$F(t) = 6 \sqrt{\frac{\lambda t}{\pi}} - 3\lambda t \quad (26) \quad /26/$$

For the data of Ref. /20/ analyzed in this work the difference between the values predicted by Eq. (25) and Eq.(26) was less than 10^{-4} , as we have found by means of computer calculations. Let us now consider the surface effects. The adsorbtion and desorbtion surface effects may generally be described with a first order law. In this case the tritium conservation equation at the particle surface is:

$$\frac{dN}{dt} = N_D(t) + k_1 p_{T_2O} - k_2 N \quad (27)$$

where: N = number of tritium atoms at the surface
 $N_D(t)$ = number of tritium atoms per unit time which reach the surface by diffusion at the time t
 k_1 = adsorbtion rate constant
 k_2 = desorbtion rate constant
 p_{T_2O} = T_2O partial pressure in the surrounding helium atmosphere

The tritium conservation equation in the particle is:

$$\frac{4}{3} \pi R^3 (c_o - \bar{c}(t)) = \int_0^t N_D(t) dt \quad (28)$$

Replacing Eq.(24) and Eq.(26) in Eq.(28) and differentiating in respect of time one obtains:

$$N_D(t) = 4\pi R^3 (c_o - c_f) \left(\sqrt{\frac{\lambda}{\pi t}} - \lambda \right) \quad (29)$$

Replacing Eq.(29) in Eq.(27) one finally obtains:

$$\frac{dN}{dt} = 4\pi R^3 (c_o - c_f) \left(\sqrt{\frac{\lambda}{\pi t}} - \lambda \right) + k_1 p_{T_2O} - k_2 N \quad (30)$$

The solution of Eq.(30) with the initial condition $N=0$ for $t=0$, is:

$$N = 4\pi R^3 (c_o - c_f) \left[\sqrt{\frac{\lambda}{\pi}} e^{-k_2 t} \int_0^t \frac{e^{k_2 t}}{\sqrt{t}} dt - \frac{\lambda}{k_2} (1 - e^{-k_2 t}) \right] + \frac{k_1 p_{T_2O}}{k_2} (1 - e^{-k_2 t}) \quad (31)$$

Thus, taking account of the tritium inventory adsorbed on the surface, one has:

$$\begin{aligned} F(t) &= 6\sqrt{\frac{\lambda t}{\pi}} - 3\lambda t - \frac{N}{\frac{4\pi R^3}{3} (c_o - c_f)} = \\ &= 6\sqrt{\frac{\lambda t}{\pi}} - 3\lambda t - 3 \left[\sqrt{\frac{\lambda}{\pi}} e^{-k_2 t} \int_0^t \frac{e^{k_2 t}}{\sqrt{t}} dt - \frac{\lambda}{k_2} (1 - e^{-k_2 t}) \right] - \frac{3 p_{T_2O} (1 - e^{-k_2 t})}{4\pi R^3 (c_o - c_f)} \frac{k_1}{k_2} \end{aligned} \quad (32)$$

With $u = \sqrt{k_2 t}$, Eq.(32) becomes:

$$\Delta F(t) = \phi(u) - \frac{3 p_{T_2O} (1 - e^{-u^2})}{4\pi R^3 (c_o - c_f)} \frac{k_1}{k_2} \quad (33)$$

where

$$\begin{aligned} \Delta F(t) &= F(t) - 6\sqrt{\frac{\lambda t}{\pi}} + 3\lambda t \\ \phi(u) &= 3 \frac{\lambda}{k_2} (1 - e^{-u^2}) - 6\sqrt{\frac{\lambda}{\pi k_2}} e^{-u^2} \cdot \int_0^u e^{u^2} du \end{aligned}$$

In Eq.(33) the term $\Delta F(t)$ is the contribution of the tritium retained on the particle surface to the fractional release, $\phi(u)$ is the fractional release due to the desorption effect and the third

term of Eq.(33) is the contribution to the fractional release due to adsorption at the particle surface.

Table V and VI show some of the data from Ref. /20/. The values for t and $F(t)$ have been obtained from the diagrams of Fig.4 and 6 of the paper, the values for D and R from Table 2 of the paper. The data of Table V have been obtained with dry helium, i.e. for $p_{T_2O} \approx 0$ and the adsorption term of Eq. (33) is negligible. Let us now try to estimate the desorption term $\phi(u)$. The desorption rate constant has been measured for THO from Li_2O /27/ and T_2O from Li_2O /28/. In both cases the constant k_2 is a function of temperature. For THO Ref. /27/ gives the equation:

$$k_2 = 1.6 \times 10^7 e^{-\frac{15458}{T}} \quad (\text{sec}^{-1}) \quad (34)$$

For T_2O we have obtained from the data of Ref. /28/ the following equation:

$$k_2 = 1.492 \times 10^8 e^{-\frac{17801}{T}} \quad (\text{sec}^{-1}) \quad (35)$$

For $T=800^\circ\text{C}=1073 \text{ K}$, the average temperature of the experiments of Ref. /20/, Eq.(34) and Eq.(35) give values of k_2 equal to 8.86 sec^{-1} and 9.30 sec^{-1} respectively. Thus for the times considered in Tables V and VI one has that

$$e^{-k_2 t} = e^{-u^2} \ll 1 \quad (36)$$

Furthermore it can be shown that

$$e^{-u^2} \cdot \int_0^u e^{u^2} du < 1 \quad /11/ \quad (37)$$

Thus, for the data of Table V and VI, it can be written (see Eq.(33)):

$$|\phi(u)| < 6 \sqrt{\frac{\lambda}{\pi k_2}} \approx 6 \sqrt{\frac{10^{-5}}{\pi \times 9}} = 0.004 \quad (38)$$

The term $3 \frac{\lambda}{k_2}$ being much smaller than the term $6 \sqrt{\lambda/\pi k_2}$. This rough estimation of $\phi(u)$ has been confirmed by more precise computer calculations.

I.e. in presence of dry helium, where adsorption does not play a role, the contribution of desorption to the fractional tritium release is negligible and the tritium release rate is diffusion controlled. This of course is not true anymore for extremely small particles, when λ becomes of the same order of magnitude as k_2 . For instance for $R \leq 10^{-4}$ cm, $\lambda \geq \frac{10^{-7}}{10^{-8}} = 10 \text{ sec}^{-1}$ and desorption becomes the rate controlling step.

The values of $\Delta F(t)$ of Table V are not exactly equal to zero due to experimental error and the error in reading the diagrams of Fig.4 and 6, however not due to the effect of desorption, because $|\phi(u)|$ is much smaller than $|\Delta F(t)|$, furthermore $\phi(u)$ should be negative while most of the $\Delta F(t)$ values of Table V are positive.

The values of $\Delta F(t)$ of Table VI are negative for low values of t , and rather large and positive for high values of t . The term due to adsorption (see Eq.(33)) is negative and this could explain the behaviour for low t values, however the data for large t values cannot be explained by adsorption⁺. A plausible explanation of the $\Delta F(t)$ values of Table VI could be the following. At the beginning, when the number N of tritium atoms at the particle surface is still relatively small, adsorption plays the dominant role and the $\Delta F(t)$ values are negative. When, however, the number of tritium atoms at the surface is higher, an effective isotope exchange between the tritium adsorbed at the surface and the hydrogen of the moisture of the sweep helium gas takes place. This exchange increases the rate of release of tritium and thus makes $\Delta F(t)$ more positive. An isotope exchange with the tritium dissolved in the particle is probably much less effective, because

⁺ Computer calculations have been performed of $\Delta F(t)$ for λ values differing up to 25% of the values given in Table VI. This scatter is considered to be the experimental error of the data of Ref./20/ and has been obtained from Fig.5 of Ref./20/. Even in this case $\Delta F(t)$ was never negative for all values of t .

it must occur via a backdiffusion of hydrogen in the particle and because the tritium inventory by solution is smaller than the inventory due to diffusion (see Section 3.2).

A determination of the adsorption rate constant k_1 is therefore only possible if one maintains a well defined T_2O partial pressure in the helium sweep flow. In this case one would expect systematically negative values for $\Delta F(t)$ and k_1 could be calculated with the equation:

$$k_1 = -\Delta F(t) k_2 \frac{3 p_{T_2O}}{4\pi R^3 (c_o - c_f)} \quad (39)$$

obtained from Eq.(33) with the conditions of Eqs. (36), (37), (38). k_2 may be calculated from Eq.(35) for T_2O and Li_2O .

Once k_1 is known the tritium inventory due to adsorption on the particle surface during reactor operation may be estimated. Indeed in steady state blanket operation in the reactor one has:

$$N_D(t) = \dot{m}_p = \text{tritium production rate in the particle} \quad (40)$$
$$\frac{dN}{dt} = 0$$

Thus Eq. (27) becomes:

$$N = \frac{\dot{m}_p + k_1 p_{T_2O}}{k_2} \quad (41)$$

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Table I: Main data of a helium cooled ceramic blanket with lead multiplier and poloidal pressure tube arrangement /4/

Total heat output = 620 MWth

First wall thickness: 10 mm (austenitic stainless steel)

Lead multiplier thickness: 107 mm

Helium coolant tubes, embedded in the lead: o.d. 29 mm, i.d. 27 mm

Pressure tubes containing the breeder material:

- outboard section: 3 rows of tubes with o.d. 150 mm, i.d. 142 mm
- inboard section: 2 rows of tubes with o.d. 125 mm, i.d. 119 mm

The outboard and inboard pressure tubes contain a perforated cylindrical calandria of 130 mm and 105 mm o.d. respectively. Cylindrical tubes are provided in the calandria for the coolant helium with an inner diameter of 12 mm and an average pitch of 34 mm.

First wall-lead section:

- helium inlet temperature = 80°C
- helium outlet temperature = 180°C
- helium inlet pressure = 80.7 bar
- helium outlet pressure = 79.3 bar
- first wall maximum temperature = 390°C
- lead maximum temperature = 295°C

Breeder section:

- helium inlet temperature = 180°C
- helium outlet temperature (average) = 380°C
- helium inlet pressure = 79.3 bar
- helium outlet pressure = 77.6 bar
- breeder material min. temperature = 235°C
- breeder material max. temperature = 750°C
- percentage of breeder material below 320°C = 35%

The breeder material is contained in the perforated calandria and it is in form of Li_4SiO_4 pebbles of 2 mm in diameter. The total pumping power required by the cooling helium in the first wall-lead and in the breeder sections is 3% of the extracted heat output.

Table II: Tritium losses for a helium purge flow at 80 bar
(poloidal solution).

$M = 0.26 \text{ kg/sec}$

$v = 1.75 \text{ cm/sec}$

helium residence time in blanket = $600 : 1.75 = 340 \text{ sec}$

tritium inventory in helium purge system = $0.34 \text{ g} = 0.34 \times 10^4 \text{ curie}$

tritium partial pressure in purge system = $19 \text{ pascal} = 0.144 \text{ torr}$

tritium losses by leakage from purge system to primary circuit =
 $0.9 \div 1.8 \text{ curie/d}$

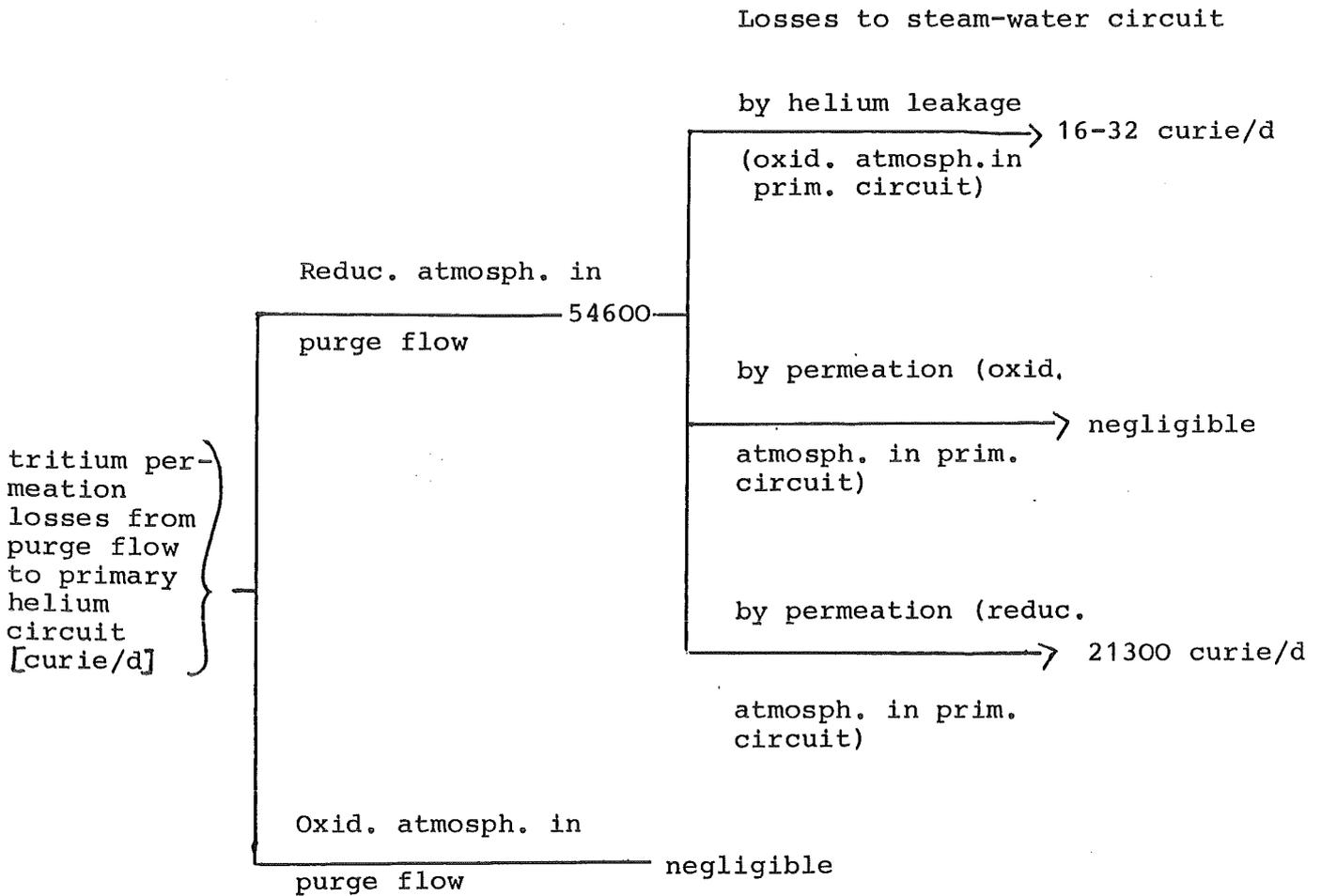


Table III: Tritium losses for a helium purge flow at 1 bar.

$M = 0.26 \text{ kg/sec}$

$v = 140 \text{ cm/sec}$

helium residence time in blanket = $600 : 140 = 4.3 \text{ sec}$

tritium inventory in helium purge system = $4.4 \times 10^{-3} \text{ g} = 44 \text{ curie}$

tritium partial pressure in purge system = $0.24 \text{ pascal} =$
 $1.82 \times 10^{-3} \text{ torr}$

tritium losses by leakage from purge flow system to primary
 circuit = $0.01 - 0.02 \text{ curie/d}$

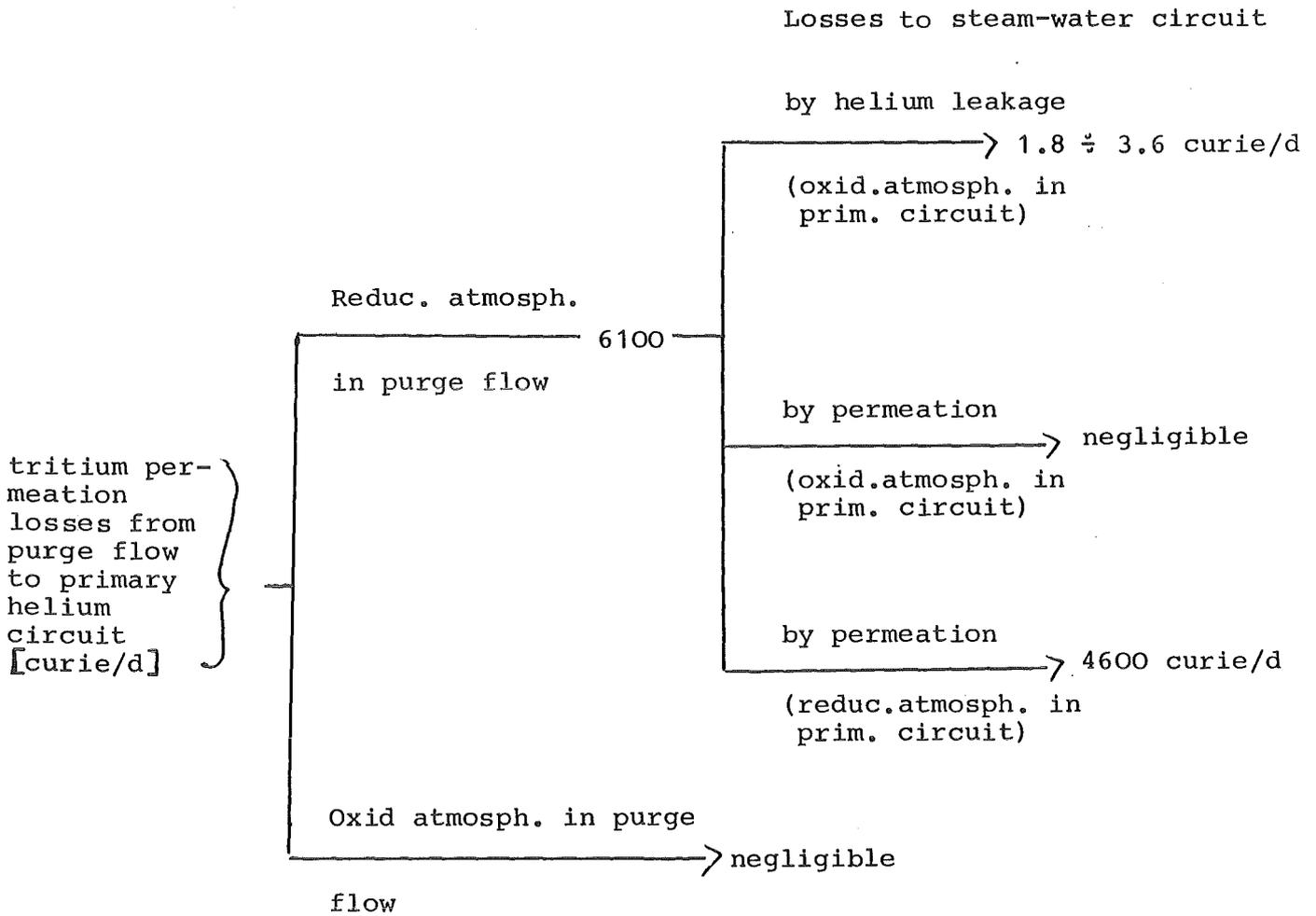


Table IV: LiOT vapor pressure as a function of temperature and of T₂O partial pressure (based on data of Ref. /25/).

P _{T₂O} (pascal)	P _{LiOT} (pascal)	
	T = 400°C	T = 850°C
19	7.6×10 ⁻⁶	1.05
12	6.0×10 ⁻⁶	0.83
0.24	0.85×10 ⁻⁶	0.12

Table V : Tritium fractional release data from Ref./20/. Li_2O . Dry helium

Fig.4, Run 1, $\lambda=1.14 \times 10^{-7}/0.113^2 = 0.8928 \times 10^{-5} \text{ sec}^{-1}$

ln t	t/sec/	F(t)	$\Delta F(t)$	$\Delta F(t)\%$
6.855	948	0.3	0.01395	4.65
7.50	1808	0.4	0.01833	4.58
8.082	3236	0.5	0.01127	2.25
8.544	5136	0.6	0.01266	2.11
8.953	7731	0.7	0.01769	2.53
9.390	11968	0.8	0.01399	1.75
9.883	19594	0.9	0.00892	0.99

Fig.4, Run 2, $\lambda=2.91 \times 10^{-7}/0.127^2 = 1.804 \times 10^{-5} \text{ sec}^{-1}$

ln t	t/sec	F(t)	$\Delta F(t)$	$\Delta F(t)\%$
6.73	837	0.4	0.02933	7.33
7.365	1580	0.5	0.01400	2.80
7.856	2581	0.6	0.009246	1.54
8.30	4024	0.7	0.00573	0.82
8.781	6509	0.8	-0.00769	-0.96
9.30	10938	0.9	-0.01169	-1.30

Fig. 4, Run 7, $\lambda=2.77 \times 10^{-8}/0.089^2=3.497 \times 10^{-6} \text{ sec}^{-1}$

ln t	t/sec	F(t)	$\Delta F(t)$	$\Delta F(t)\%$
7.778	2387	0.3	0.01576	5.25
8.434	4601	0.4	0.01888	4.72
9.007	8160	0.5	0.01377	2.76
9.45	12708	0.6	0.01971	3.28
9.87	19341	0.7	0.02254	3.22
10.3	29733	0.8	0.02038	2.55
10.78	48050	0.9	0.01647	1.83

Table VI: Tritium fractional release data from Ref./20/, Li₂O.
Helium with ~ 1 vpm H₂O

Fig.6, Curve 1, T=800°C, R=0.116 cm/24/, D=2.791x10⁻⁷ cm²/sec /20/,
λ=2.074 x 10⁻⁵sec⁻¹

ln t	t/sec/	F(t)	ΔF(t)	ΔF(t)%
6.4	602	0.3	-0.04082	-13.61
6.86	953	0.4	-0.01665	-4.16
7.25	1408	0.5	0.00910	1.82
7.56	1920	0.6	0.04392	7.32
7.87	2618	0.7	0.07406	10.58
8.18	3569	0.8	0.10104	12.63
8.526	5044	0.9	0.11892	13.22

Fig.6, Curve 2, T=850°C, R=0.151 cm /24/, D=4.197 x 10⁻⁷ cm²/sec /20/,
λ=1.841 x 10⁻⁵sec⁻¹

ln t	t/sec	F(t)	ΔF(t)	ΔF(t)%
6.48	652	0.3	-0.03484	-11.61
6.98	1075	0.4	-0.01682	-4.21
7.43	1686	0.5	-0.00325	-0.65
7.77	2368	0.6	0.02402	4.00
8.10	3294	0.7	0.04835	6.91
8.44	4629	0.8	0.06749	8.44
8.80	6634	0.9	0.08341	9.27

Fig.6, Curve 3, T=750°C, R=0.149 cm /24/, D=1.784 x 10⁻⁷ cm²/sec /20/,
λ=8.034 x 10⁻⁶sec⁻¹

ln t	t/sec	F(t)	ΔF(t)	ΔF(t)%
7.21	1353	0.3	-0.02033	-6.78
7.71	2231	0.4	0.00056	0.14
8.15	3463	0.5	0.01882	3.76
8.48	4817	0.6	0.05016	8.36
8.82	6768	0.7	0.07375	10.54
9.17	9605	0.8	0.09113	11.39
9.60	14765	0.9	0.08996	10.00

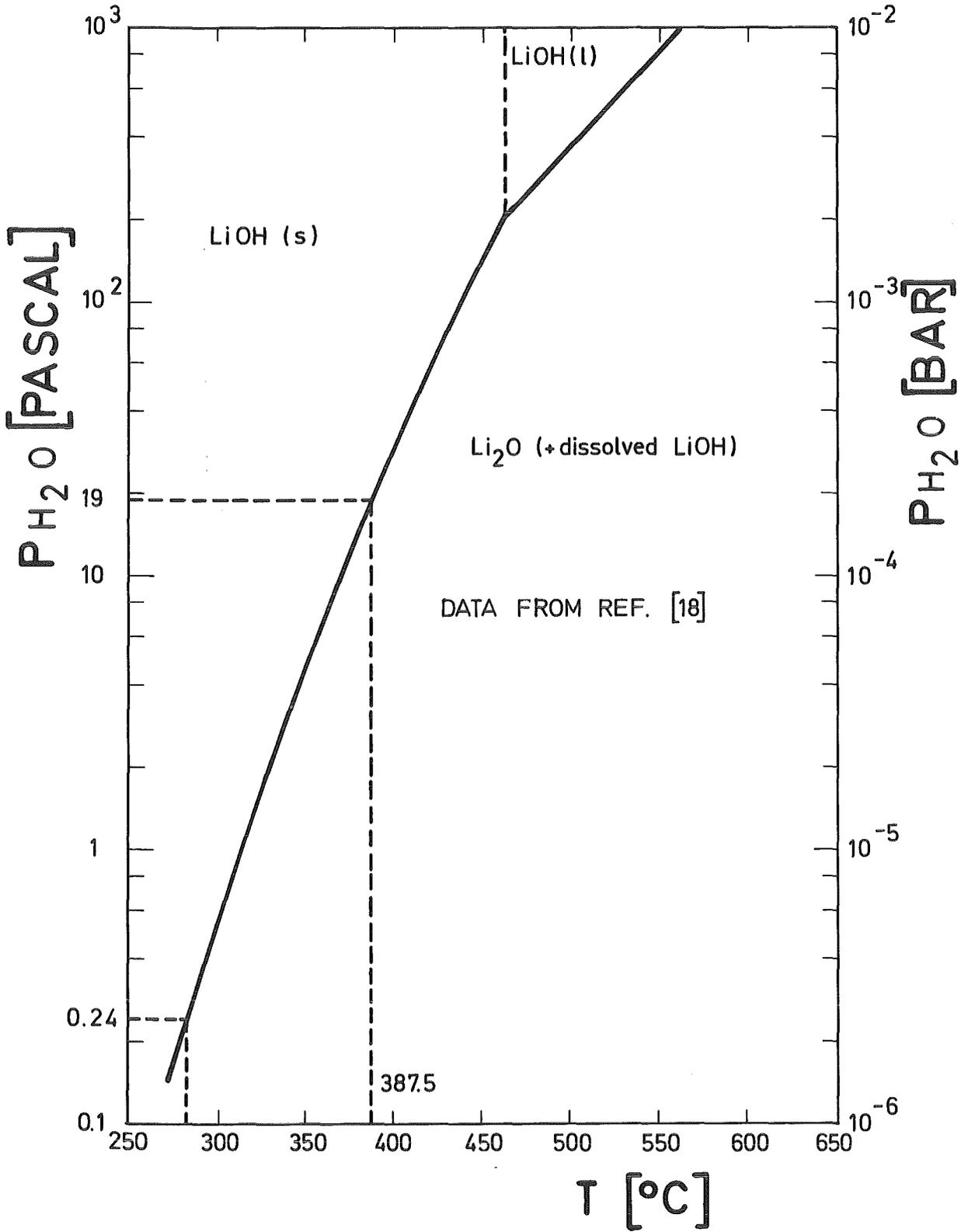


Fig.1: Partial pressure of H₂O above the diphasic Li₂O(solid)-LiOH (solid, liquid) system as a function of temperature /18/.