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TRITIUM BEHAVIOUR IN CERAMIC BREEDER BLANKETS

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Abstract

Tritium release from the candidate ceramic materials, Li_2O, $LiAlO_2$, Li_2SiO_3, Li_4SiO_4 and Li_2ZrO_3, is being investigated in many blanket programs. Factors that affect tritium release from the ceramic into the helium sweep gas stream include operating temperature, ceramic microstructure, tritium transport and solubility in the solid. A review is presented of the material properties studied and of the irradiation programs and the results are summarized. The ceramic breeder blanket concept is briefly reviewed.

1. INTRODUCTION

A breeder blanket for a DT fusion reactor has two main purposes: to remove the heat produced in the blanket by the fusion reaction neutrons and to breed the tritium required to sustain the reaction. To meet the latter objective, lithium is included as a component of the blanket material because of its high neutron absorption cross-section. Tritium will be generated from neutron capture by ^6Li and ^7Li.

$$^6Li + n \rightarrow {}^3H + {}^4He$$
$$^7Li + n \rightarrow {}^3H + {}^4He + n$$

Solid breeder blankets and liquid breeder blankets are the two generic classes of blanket concepts. Design concepts within each class involve a variety of breeder material, coolant, and configuration choices. Recent fusion breeder blanket review studies include the US Blanket Comparison and Selection Study (BCSS) [1], a Canadian review [2], the FINESSE study [3] and a follow-up technical study to FINESSE [4] which examined selected technical problems for the very near-term experiments (< 3 years) and began development of analytical and theoretical models for nuclear components.

This paper focuses on solid ceramic breeder blankets, on issues specifically relating to tritium release and recovery, and on the importance of these issues to the advancement of blanket designs. Material properties and blanket design considerations are briefly reviewed.

A number of ceramic breeder blanket options are being tested and evaluated. The major compounds presently under study include Li_2O and the ternary oxides, $LiAlO_2$, Li_2SiO_3, Li_4SiO_4 , and Li_2ZrO_3. The testing and evaluation includes development of fabrication techniques, thermomechanical behaviour studies, material property measurements, and irradiation and tritium release behaviour.

2. BLANKET DESIGNS

As well as evaluating the breeder material, design of a blanket must consider structural materials, coolant, addition of a neutron multiplier, and optimization of the size and configuration for the specific operating requirements. In the present design concepts, the ceramic material is contained in tubes or canisters and tritium release is accomplished by flow of helium sweep gas through purge channels in the solid. Evolved tritium is carried by the sweep gas to an extraction facility. The ceramic breeder must be maintained within upper and lower temperature limits and this is complicated by nuclear heat deposition which is high near the front wall and falls rapidly beyond it. The design for cooling must accommodate this heat distribution. The coolant may be external to the breeder container (Breeder in Tube, BIT) or the coolant tubes may penetrate through the breeder volume (Breeder out of Tube, BOT) [5]. Both water-cooled and helium-cooled designs are considered. To minimize cracking by thermal stresses, which could affect the thermal conductance between the breeder and the coolant, the breeder material may be in the shape of pebbles or small spheres. But monolithic material can also be used in the form of larger, solid pellets or blocks if it possesses high thermal shock resistance. To achieve a high tritium breeding ratio (tritium atoms produced > tritium atoms burned), a neutron multiplier material such as beryllium may be necessary. This introduces other complexities as the structural and blanket material must be compatible with it.

The complexity of the design of a solid breeder blanket is shown in Figure 1, which illustrates a helium-cooled BOT concept. As explained by Vetter [5], this outboard blanket is composed of self-supporting canisters containing a beryllium multiplier in the form of plates and the interspaces are filled with a bed of 0.5 mm diameter Li_4SiO_4 spheres. The coolant and purge gas manifolds are arranged on the back side of the canisters, presenting a dense pack towards the front side to gain a high efficiency of breeding. The purge gas flows in a separate circuit at atmospheric pressure. The main coolant stream is guided through the cooling channels of the first wall before entering the manifold of the breeder/multiplier submodules.

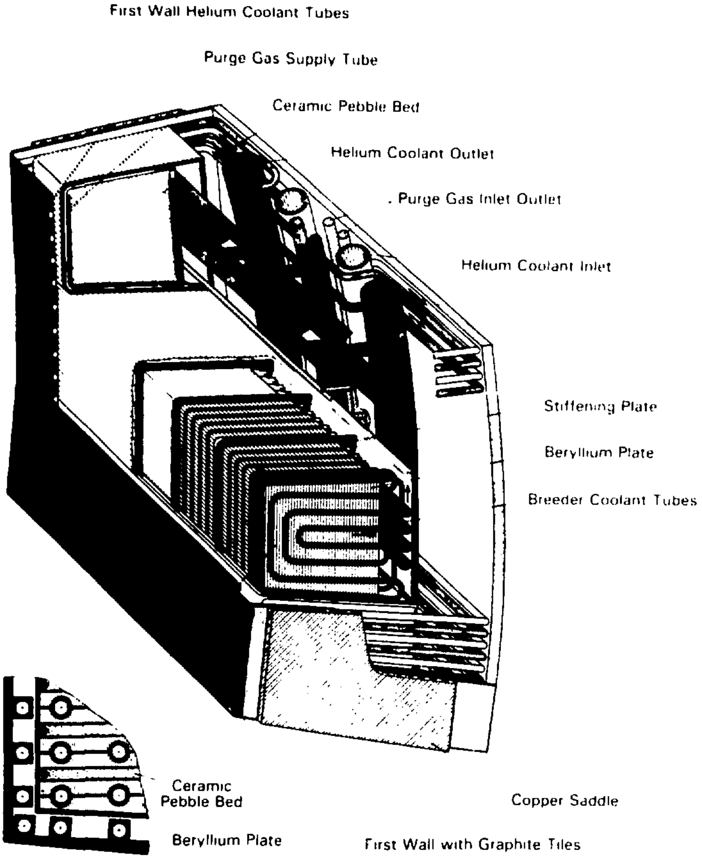

Figure 1 - Breeder Out of Tube (BOT) Blanket Concept; View of Outboard Blanket Canisters [5]

The critical issues for solid breeder blankets are summarized in Table 1.

Table 1 - Critical Issues for Solid Breeder Blankets [3]
- tritium self-sufficiency
- breeder/multiplier tritium inventory and recovery
- breeder/multiplier thermomechanical behaviour
- corrosion and mass transfer
- structural response and failure modes
- tritium permeation and processing from the blanket

3. BREEDER MATERIAL PROPERTIES

Tritium release properties cannot be considered in isolation from the material properties of the chosen ceramic compound. The operating temperature window for solid breeder blankets is limited to approximately 350 - 1000°C, depending on the material. The bounds are set to keep the temperature high enough for acceptably rapid tritium diffusion and desorption but not so high as to cause sintering and vapour transport. It is desirable for tritium release to occur at as low a temperature as possible. These solid materials all have a relatively low thermal conductivity, ~1-3 W/m.K [6], which will cause steep temperature gradients and possible breeder fragmentation. For this reason, spheres and pebbles are used in some designs, as in Figure 1, rather than the monolithic solids. A comparison of characteristics of the main breeder candidate ceramics is given in Table 2 [7]. The low temperature limit is established from tritium inventory considerations and the upper limit is dictated by thermal stability and chemical compatibility. The lower temperature limit will be affected by the microstructure of the ceramic material. From the experimental results given in the table, upper temperature limits of 800°C for Li_2O and Li_4SiO_4, 950°C for Li_2ZrO_3, and 1000°C for $LiAlO_2$ are suggested.

Characteristics	$LiAlO_2$	Li_4SiO_4	Li_2ZrO_3	Li_2O	Source of Experimental Data
Melting point in °C	1610	1250	1615	1430	
Li content (g cm^{-3})	0.27	0.54	0.38	0.93	
Thermal conductivity at 600°C, 80% TD (W/m•K)	2.6	2	1.4	3.5	
Fragmentation	low	very high	no	medium	F U B R
Swelling (900°C, 297 FPD)	<0.5%	1.5%	<0.7%	8%	
Grain growth					
100 FPD, 700°C	no	no	no	3.5 μm → 7 μm	1A
900°C	no	1 μm → 2 μm	no	3.5 μm → 17 μm	
Lithium mass transfer (900°C)	no	0.16 at %	no	0.9 at %	
Temperature corresponding to 1 day T_2 residence time	450°C	390°C	310°C	320°C	LILA LISA MOZART
Grain size and origin	0.4 μ CEA	21 μ KfK	1 μ U.S	16 μ JAERI	

Table 2 - Summary of Material Characteristics [7]

Other material parameters, such as grain size, pore size, form and impurity content, will also affect the tritium release performance of a particular ceramic compound in a blanket concept. These parameters may be influenced by radiation damage and/or interaction of the breeder material with the structural and neutron multiplier material, which has the potential to affect tritium release properties.

4. TRITIUM RELEASE AND INVENTORY

To fully predict tritium behaviour in solid breeders requires understanding of tritium transport, retention and chemical form in the breeder and in the multiplier material under the influence of the fusion environment [3]. Factors that affect tritium release include the microstructure of the breeder material, the operating temperature, and tritium transport and solubility in the solid. Analysis of the tritium release process has considered the following transport mechanisms [4, 8]:

- (i) intra-granular diffusion to the solid breeder grain boundary;
- (ii) diffusion along the grain boundary (including the contribution of grain boundary porosity);
- (iii) desorption from the free surface; and
- (iv) transport in the purge flow.

Intra-granular diffusion and surface desorption are generally considered to be the rate-limiting steps in the release process.

Tritium retention, or the tritium inventory in the blanket material, is assumed to be the sum of three components; a diffusive inventory, a solubility inventory, and a surface adsorption inventory. The diffusive inventory is a function of the diffusion coefficient of the particular material, the grain size and the operating temperature. A significant amount of diffusion data for several materials is available. Some of these data have been determined from specific diffusivity measurements, while others have been obtained from tritium release from irradiated samples. Values for specific materials, though, are spread over several orders of magnitude. In obtaining diffusion coefficients from tritium release studies, it is first essential to establish that the tritium release process is controlled by diffusion and not by some other release mechanism. Also, determining a diffusion coefficient from tritium release from powder and pellets, whose surface areas are high when compared to single crystals, is complicated by the fact that tritium release may be affected by a surface reaction. Guggi [9] and Tanifuji *et al.* [10] have measured the diffusivity of tritium in single-crystal Li_2O and good agreement was obtained between the two (0.3 to 1.1×10^{-7} cm^2/s at 873 K)., Kudo *et al.* [11] have measured the diffusion coefficient for tritium in tritium-release studies in crystalline powders of Li_2O, $LiAlO_2$, Li_2SiO_3, Li_2ZrO_3 and Li_8ZrO_6. While their value for Li_2O is approximately an order of magnitude less than Guggi's at 873 K, Figure 2, taken from reference 11, gives an indication of the relative diffusivity for these materials. It should be noted that Kudo *et al.* also indicate a difference in the diffusion coefficient between irradiated and unirradiated Li_2O, with the diffusivity being higher in the neutron-irradiated sample. The effects of irradiation, if any, on the determination of a diffusion coefficient need to be investigated further.

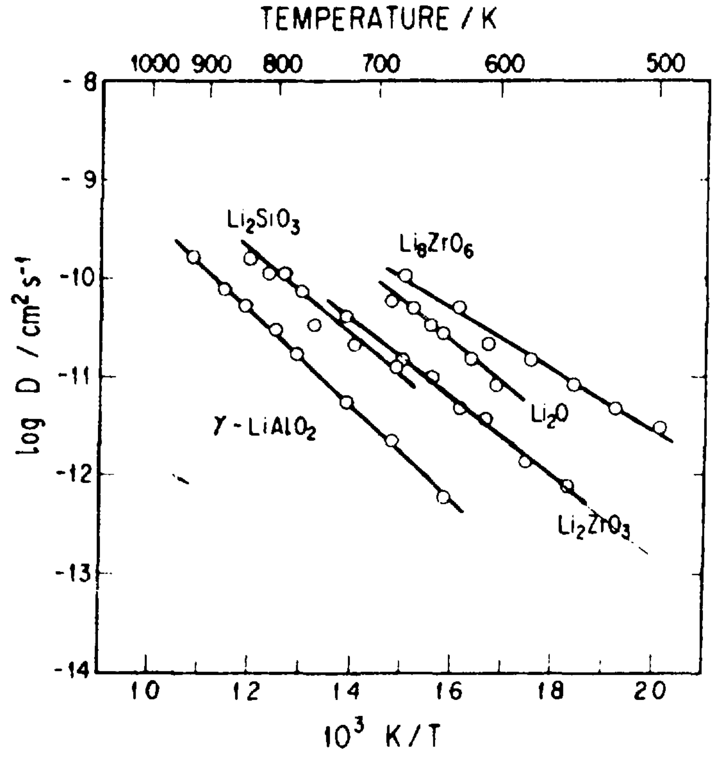

Figure 2 - Diffusion Coefficients for Candidate Breeder Materials [11]

Phase equilibria and tritium solubility are important parameters for their effects on tritium inventory, but only limited thermodynamic studies have been carried out on the compounds of interest. The Li_2O/H_2O system has been studied extensively [12, 13] and the solubility of the hydroxide (tritoxide) determined as a function of the moisture partial pressure and temperature in a single phase system. The solubility of LiOH was found to increase with temperature at a constant partial pressure of water, making the LiOH-Li_2O system thermodynamically non-ideal. Experimental work was also carried out by Tetenbaum and Johnson [13] to determine the partial pressure of water vapor required to precipitate LiOH from Li_2O in the temperature region 573 to 894 K. To avoid precipitation of a second phase, LiOH(T), at 700 K the partial pressure of H_2O must be maintained below 71 Pa, at 900 K below 2.4 kPa. To limit tritium inventory, the blanket must not operate in a regime where LiOT is the stable phase.

Experimental studies on the solubility of OH^- in $LiAlO_2$ at 673 and 773 K as a function of the partial pressure of H_2O have been recently carried out by Fischer and Johnson [14] in ultra-pure helium sweep gas. They have extrapolated their results to reactor blanket conditions of 1 wppm $H_2O_{(g)}$ at 10^5 Pa total pressure to give 0.39 and 0.18 wppm T in $LiAlO_2$ at 673 and 773 K, respectively, due to the solubility of tritium in the form of tritoxide. The influence of H_2 added to the sweep gas on the solubility also needs to be examined. Their data also indicate that the overall adsorption/solubility process is exothermic, as a decrease was observed when the temperature was increased, in contrast to solubility data in Li_2O which was endothermic.

The solubility of HT or T_2 may also contribute to the tritium inventory. Some studies on the solubility of H_2 and D_2 in these materials have also been carried out. Kudo and Okuno [15] have investigated the solubility of H_2 in Li_2O and Glugla *et al.* H_2 solubility in Li_2SiO_3 [16]. The hydrogen solubility was observed to increase with temperature for both compounds and is of comparable magnitude within the temperature range studied. A solubility of 3.6×10^{-5} mol H/mole Li_2SiO_3 was measured at 500°C. Possible isotope effects with tritium still need to be determined.

The surface adsorption/desorption process is complex, and again only very limited thermodynamic data is available. Yoshida *et al.* [17] have studied surface adsorption of H_2O on Li_2O, and Fischer and Johnson's study [14] included the adsorption of $H_2O_{(g)}$ on $LiAlO_2$ and the kinetics of $H_2O_{(g)}$ evolution. While the study by Fischer and Johnson was limited to 673 and 773 K in a ultra-pure helium sweep gas environment, the adsorption isotherms obtained did indicate more than one kind of surface site for adsorption, each with distinctive adsorption energies. This knowledge is important for understanding the adsorption/desorption process, and for development of a tritium release model.

For both safety and economic reasons, tritium inventory in an operating blanket must be minimized. With limited experimental data, however, there is a large uncertainty in this inventory level. Along with the material properties, the breeder chemical environment, the effects of radiation-trapping in the breeder and of the addition of a beryllium multiplier material on tritium inventory need to be addressed.

The breeder chemical environment, and in particular the oxygen activity (in effect, the O_2 partial pressure), also affects the chemical form of the tritium that is released and/or recovered from the blanket. The tritium could be present in the oxide (HTO) or elemental (HT) form. Purge gas chemistry, breeder material, multiplier and structural materials, and operating temperature will all influence oxygen activity around the breeder surface. Thermodynamic calculations by Fischer and Johnson [18] have shown the importance of oxygen activity on the chemical form obtained from Li_2O. An example is given in Figure 3, where the calculated amounts of HTO and HT are shown as a function of oxygen activity for different temperatures. Knowing the form of the tritium will be important in an operating breeder blanket both for safety reasons (HTO is approximately 10^4 times more toxic that HT) and recovery process design.

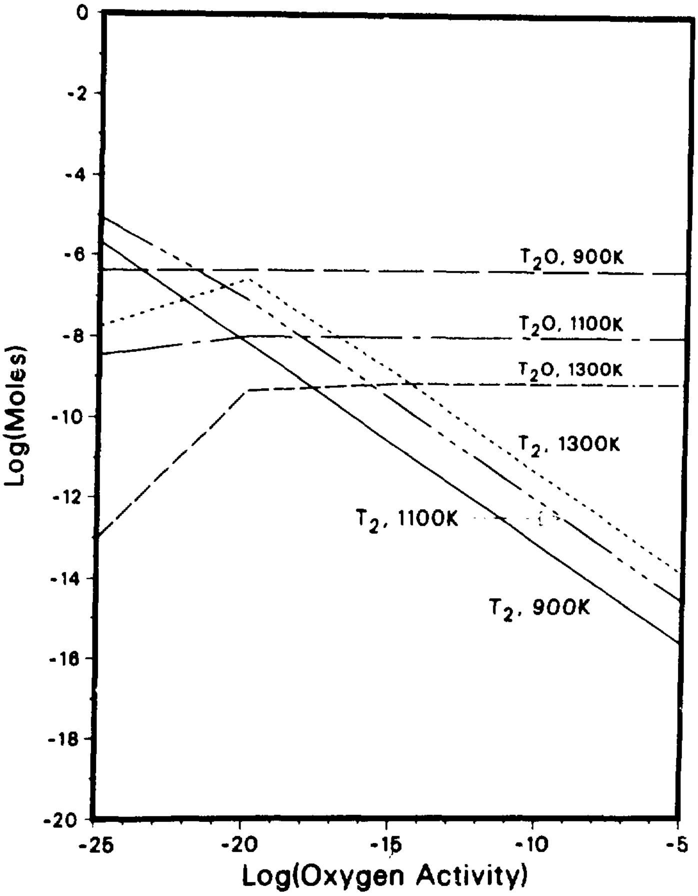

Figure 3 - Quantities of Gas Phase Species (T_2 and T_2O) at 900, 1100 and 1300 K as a Function of Oxygen Activity in a Protium Free System [18]

5. TRITIUM EXTRACTION

Purge gas composition, amount of H_2 added, tritium levels, HTO/HT ratio and breeder impurities will all influence the choice and design of an extraction system, as well as the design and operating requirements of the blanket itself. While little experimental work has been done in this area, the concepts presented are based on established process technology. For low-pressure purge-gas purification, catalytic oxidation followed by cryogenic adsorption has been suggested [19]. On regeneration of the cryoadsorber, the HTO can be further processed by electrolysis, with the hydrogen isotopes stream being fed to an isotope separation system for final recovery of the tritium. This process is not as attractive for a purge gas of high pressure and large flow because of adverse economics for operation of large cryoadsorbers. Dombra *et al.* [20] have developed a concept for a system with high pressure (8 MPa) and large flow (7 m^3/s), the requirements in a NET-blanket design. This concept uses catalytic oxidation followed by adsorption of HTO on molecular sieve driers. After regeneration of the driers, the tritium is recovered using a catalytic isotopic exchange process, followed by cryogenic distillation. While not being directly tested for tritium extraction from the blanket purge gas, many of these technologies are being investigated and demonstrated for other parts of the fusion cycle.

6. IRRADIATION TESTING

Two types of ceramic-blanket irradiation tests are performed to examine tritium release: (i) unvented capsule tests in which tritium-release information is obtained via post-irradiation annealing, and (ii) vented (or *in-situ*) tests which permit on-line monitoring of the tritium release as the irradiation progresses.

6.1 Unvented Tests

The unvented irradiation tests are particularly useful as scoping studies for new materials, examining the effect of material characteristics on tritium release, investigating the effect of the chemical environment and developing tritium release mechanisms [21, 22, 23]. Generally, short irradiations are carried out, in the order of a few hours to a few days, using small samples (<100 mg) and generating only up to the order of 10^7Bq of tritium per sample. In these tests, tritium release information is obtained after the irradiation is complete and the capsule has been removed from the reactor. Operation of the tritium recovery and analysis system generally involves heating the sample in a flowing sweep gas, which carries the released tritium to an on-line ionization chamber or proportional counter, and/or bubblers for removal of the tritium and subsequent analysis by liquid scintillation counting. Because the form of the tritium recovered, i.e., oxidized (HTO) or reduced (HT), can vary with the capsule material and sweep gas composition, components or equipment to reduce and/or oxidize the tritium may be included in the analysis system. Table 3, taken from reference 21, summarizes data on the effect of the oxygen activity of the system on the form of the tritium release in tests from the CREATE-series (Chalk River Experiment to Assess Tritium Emission). Generally, with either He-1% H_2 sweep gas or a stainless steel extraction vessel, the oxygen activity was very low and the tritium was recovered primarily as HT. Primarily HTO was recovered when the oxygen activity was raised by use of pure He sweep gas or extraction vessels made from materials relatively inert compared to stainless steel. Because of the small amounts of tritium generated in these tests, particular attention must be paid to material and system impurities and their effect on the data obtained.

Figure 4 shows the effect of changing the sweep gas composition on tritium release from $LiAlO_2$ samples in post-irradiation tests by Roth *et al.* [24]. The effect of adding H_2 to He sweep gas to increase the tritium release rate is clearly noted. Adding 0.1% CO also had an effect, but a much weaker one.

		He Sweep Gas		He-1% H_2 Sweep Gas	
Ceramic	Extraction Vessel	Predominant Form	% HTO	Predominant Form	% HT
$LiAlO_2$ (~0.05-0.3 μm grain size)	Quartz	HTO	67-87 (2)*	HT	63-81 (2)
	Stainless Steel	HT	5-20 (2)	HT	87 (1)
	Nickel	HTO	70 (1)		
$LiAlO_2$ (0.2-1 μm grain size)	Quartz	HTO	51 (1)	HT	86-88 (3)
$LiAlO_2$ (0.1-1 μm grain size)	Quartz	**	**	HT	73-90 (5)
$LiAlO_2$ (2-10 μm grain size)	Quartz	**	**	HT	47-50 (2)
Li_2O (4-15 μm grain size, some 30-40 μm)	Quartz	HTO	58 (1)	HT	80 (1)
	Inconel 600	HTO	64 (1)	HT	68-70 (2)
	Stainless Steel	HT	46-53 (2)	HT	81 (1)

* Number in brackets is the number of samples tested under the given conditions
** Material was not tested under these conditions.

Table 3 - Influence of Sweep Gas Composition and Extraction Vessel Material on the Chemical Form of Tritium Recovered [21]

The FUBR-1A and -1B closed-capsule irradiations [25, 26] were prolonged, up to 600 full power days, and were carried out to obtain information on the thermal and mechanical performance and tritium retention of various materials in a fast reactor environment, which allows more homogeneous, higher ^6Li burn-ups. As well as achieving moderate to high lithium burn-up levels, large diameter samples could be irradiated under large temperature gradients. Tritium retention under these irradiation conditions can also be examined in post-irradiation studies to determine the extent, if any, of tritium trapping due to radiation damage [27].

6.2 Vented Tests

Vented capsule tests permit continuous on-line monitoring of the tritium release from the ceramic during the irradiation, by passing a sweep gas around or through the ceramic and into an analysis train. Table 4 is a partial list of a variety of tests of this type. These tests allow one to measure important fundamental parameters (diffusion, desorption, and tritium residence time) so that the behaviour of other blanket assemblies can be inferred. As the tritium recovery process will be similar in an operating fusion blanket, this type of experiment attempts to model a miniature element of a blanket.

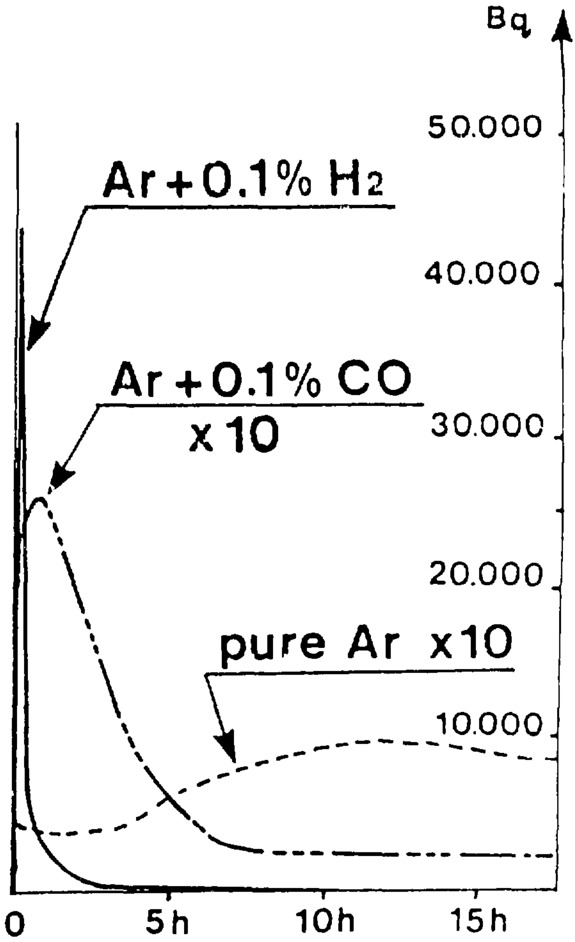

Figure 4 - Effect of Sweep Gas Composition on Tritium Release from $LiAlO_2$ in Post-Irradiation Annealing [24]

The sweep gas flowing through the test material collects the tritium released from the lithium ceramic. It then passes through a tritium analysis system to determine the tritium release rate and the chemical form of the tritium. For a chosen material with specific characteristics, the ceramic temperature and the sweep gas composition can be varied to determine their effect on the release rate and form of tritium recovered. Other variables include flow rate and neutron flux.

A schematic of the CRITIC-I (Chalk River In-Reactor Tritium Instrumented Capsule) tritium analysis train, which allowed the determination of the tritium form, is given in Figure 5 [28]. On-line ionization chambers measured total tritium (HTO + HT) and HT only concentrations. A set of ethylene glycol bubblers following the first ionization chamber removed the HTO from the sweep gas stream. A second set of bubblers removed the HT

component, after conversion to HTO in a CuO bed. Determination of the total HTO and HT over a period of time was then obtained by analysis of the bubbler solution by liquid scintillation counting.

Test Designation	Materials	Particulars
TRIO (U.S.)	$LiAlO_2$	42 g of 9 mm annular pellets, He and He-0.1% H_2 sweep gas, temperature step-change tests over range 400-700°C, 1.48 TBq of tritium collected.
LISA, -1, -2 (Germany)	Li_2SiO_3 Li_4SiO_4 Li_2O	Material comparison, He-0.1%H_2 sweep gas, temperature step-change tests over range 350-650°C.
VOM (Japan)	(-22) Li_2O $LiAlO_2$ (-23) $LiAlO_2$ Li_4SiO_4	Material comparison. VOM-22H had 5 mm spheres with 10 µm grain size, temperature range from 300-900°C, sweep gas composition, varied from He to He-1%D_2. In VOM-23, fine-grained (0.40 µm) $LiAlO_2$, and 14 µm grain size Li_4SiO_4 were compared, He-0.1% D_2 sweep gas, temperature step-change tests over range 500 - 800°C. Similar release results obtained for the two.
LILA, -1, -2, -3 (France)	$LiAlO_2$	Effect of different textures - grain size and density, temperature step-change tests over range 400-600°C, sweep gas compositions tested - He, He-0.1%CO and He-0.1%H_2.
EXOTIC, -1, -2, -3 (Netherlands/UK/ Belgium)	(-1) Li_2SiO_3 $LiAlO_2$ (-2) Li_2SiO_3 Li_2O (-3) Li_2SiO_3 $Li_2Si_2O_5$ Li_4SiO_4 Li_2ZrO_3	Material comparison - different density, grain size, ^6Li content, single temperature over range from 400 to 600°C, He-0.1%H_2 sweep gas, vary irradiation time.
CRITIC-I (Canada)	Li_2O	Long-term irradiation, 100 g large-grained (50 µm) annular pellets, temperature step-change tests 390-800°C, vary sweep gas composition from He to He- 1%H_2, achieved 1% burnup, collected 78 TBq of tritium.
MOZART (France/US/Japan)	$LiAlO_2$ Li_2ZrO_3 Li_2O	Material comparison, investigate performance at low temperature, temperature step-change tests, pure He and He-0.1%H_2 sweep gas.

Table 4 - Summary of Irradiation Tests To Date

Most experiments have not attempted to measure the chemical form because of the difficulty in quantitatively transporting the small concentrations of HTO over the piping length. Rather, a zinc bed has been located just downstream of the irradiation capsule to reduce the tritium [29] for measurement only of the total tritium released. A ceramic electrolysis cell was used in some VOM experiments for the same purpose [30]. In CRITIC-I, the relatively high tritium concentrations (0.7 - 1.0 TBq/m^3) and the heated sweep gases lines limited this concern. The determination of the chemical form has also required that the capsule materials and sweep gas system design be chosen to reduce their influence on the form measured.

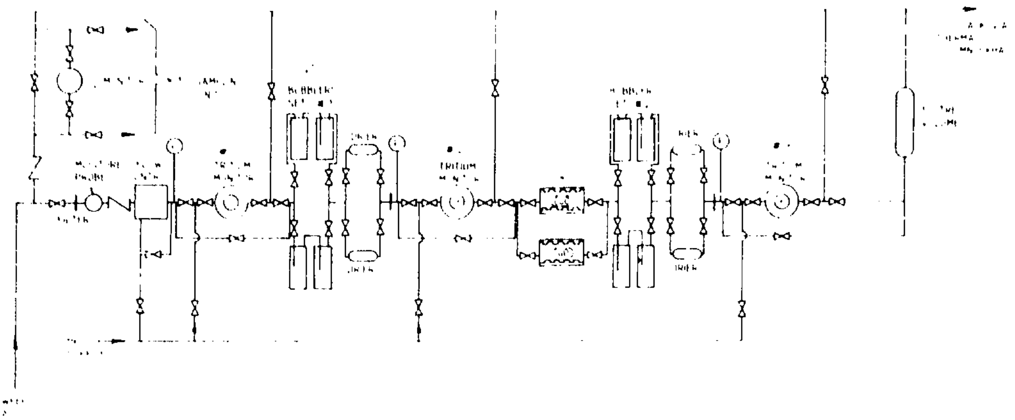

Figure 5 - CRITIC-I Tritium Analysis Train

The data required to examine diffusion and/or desorption as the rate-limiting step in the tritium release process are obtained by observing the change in the tritium release rate (from steady-state conditions) when an experimental parameter like temperature or sweep gas composition is changed. Figure 6 [28] illustrates the effect of a step-change in temperature during the CRITIC-I experiment. A temperature change from 447 to 535°C resulted in a positive tritium release peak (release greater than steady-state release) and a subsequent decrease in the tritium inventory, determined by the area under the peak. The result of this temperature step-change was also influenced by the sweep gas composition, as shown. The time constant for the tritium release peak, which is a measure of the tritium residence time in the ceramic, with He-1% H_2 sweep gas was 1.8 h compared to 21 h with He-0.1% H_2 sweep gas, indicating the positive effect of H_2 on minimizing the tritium inventory.

Analysis of the *in-situ* tritium release data from the various experiments indicates a lot of complexity in the tritium release process. While similar compounds have been irradiated in a number of tests, comparisons between tests is difficult because of different material properties, such as density, grain size, surface area, and impurities, and differences in the testing programs, temperatures, sweep gas composition, tritium production rate and lithium burn-up achieved. Tests such as EXOTIC [31] and MOZART [32] have been carried out with a number of materials of different characteristics to allow comparisons between materials under the same test conditions.

To minimize tritium inventory, the blanket conditions (especially temperature) should be chosen to achieve a tritium release rate equal to the generation rate within 24 hours. For a particular compound, the temperature at which steady-state tritium release is achieved is

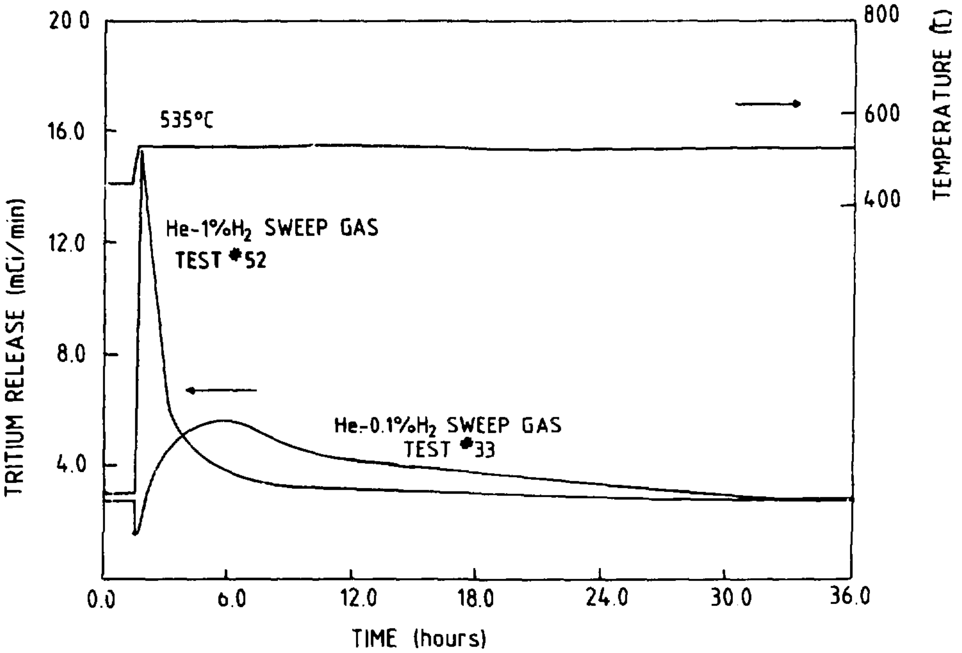

Figure 6 - Tritium Release from Li_2O (CRITIC-I) as a Function of Time Following Temperature Step-Change Test, 447 to 535°C, He-0.1%H_2 and He-1%H_2 Sweep Gases, 100 mL/min [28]

dependent on microstructural details of that material (grain size, density, etc.) and the sweep gas composition. Achieving good tritium release at a low temperature allows downward extension of the operating temperature window of a blanket while maintaining acceptable tritium inventory. In MOZART [32], an acceptable tritium release was obtained at temperatures as low as 310°C for Li_2ZrO_3. While the results from the VOM-22H, CRITIC-I and EXOTIC-2 experiments [30, 33, 34] on the irradiation of Li_2O do not compare in detail, the results do show qualitative agreement on the minimum temperature of 400 - 450°C, depending on the particular test conditions. However, the MOZART irradiation of Li_2O indicated a minimum temperature in the order of 310 - 320°C. Again, the influence of the material parameters, test conditions, and irradiation history must be taken into account. Good release was also observed in this temperature range from Li_4SiO_4 in LISA [29]. A minimum temperature for $LiAlO_2$ is very dependent on the grain size. Results from MOZART give 450°C for material with 0.4 μm grain size. The results from the various *in-situ* tests all concur on the addition of H_2 to the sweep gas enhancing the tritium release, as shorter residence times were obtained and the tritium inventories were reduced.

6.3 Modelling

Tritium release information from the irradiation testing is being used to develop release models to gain an understanding of the tritium release process and prediction of tritium release and inventory from blanket designs. The complex experimental data obtained under the various test conditions cannot be explained by simple kinetic expressions for diffusion or desorption. A relatively simple model for tritium inventory in the ceramic was developed by Briec *et al.* [35] to take into account both the bulk diffusion and surface desorption processes. If tritium solubility is assumed small, the inventory, I, in the ceramic is given by

$$I = \frac{\dot{I}R^2}{15D} + \frac{\dot{I}R}{3h} = \frac{\dot{I}R}{3h}\left\{1 + \frac{hR/D}{15}\right\} \qquad (1)$$

where $\dot{I}$ is the tritium generation rate, R the grain size, D and h are the coefficients of diffusion and mass transfer at the interface, respectively. For $hR/D < 0.5$, the second term can be neglected. While qualitative agreement was obtained with some data [28, 35], a more complicated model is required to explain results obtained over a range of conditions. More complexity is included in the tritium release model developed by Kopasz *et al.* [36, 37] which is the effect of changing the surface coverage. As the surface-fraction coverage of hydrogen and tritium is changed, the activation energy for desorption must be changed. Briec, from the results of the MOZART experiment [32], infers that, while the main mechanisms influencing tritium release are intragranular diffusion and tritium desorption at the solid-gas interface, their relative importance depends on the specific surface area and temperature of the sample. Desorption becomes controlling when the grain size and temperature decrease. This result of changing from a diffusion-controlled to desorption-controlled regime was also developed by Bertone [38]. While the release data are not yet fully understood, material parameters, the effect of impurities, particularly H_2O and CO_2 in the ceramic, the amount of H_2 and H_2O in the sweep gas, and the operating temperature have all indicated an influence on the tritium release and inventory.

6.4 Future Testing

In-situ tritium release testing to date has been carried out in a thermal neutron (mixed-spectrum) environment. To more closely approximate the fusion neutron environment (0 - 14 MeV energy range), and to help determine possible effects of fast neutrons, tritium release will be examined in a high-energy-neutron environment (1 MeV), in the BEATRIX-II (Breeder Exchange Matrix, co-ordinated by the IEA) experiment in the Fast Flux Test Facility (FFTF) in Hanford, WA, USA [39]. In Phase I, scheduled to begin 1989 September, two *in-situ* capsules containing Li_2O will be irradiated with lithium atom burnups to 4% and with higher tritium production than in previous tests, approximately 0.3 PBq during the 300 equivalent full power days of irradiation with a reference steady-state tritium partial pressure of 50 vppm. One capsule possesses the capability for temperature-change experiments, while the other exhibits a temperature gradient for evaluating the temperature stability in an engineering blanket configuration.

7. BLANKET DESIGN REQUIREMENTS

R&D studies have shown that acceptable tritium release can be obtained from lithium ceramics. However, the tritium release process is complex and influenced by a number of material properties and blanket operating requirements. The specific material, microstructure characteristics, and thermomechanical behaviour, along with tritium release characteristics, operating temperature range, and tritium inventory will need to be assessed in the context of a blanket design suitable for a fusion machine.

A review of the testing needs and experiments for solid breeder blankets by Gierszewski and Puigh [40] outlined the most important uncertainties for the use of solid breeder materials in blanket designs: (1) there is limited understanding of gas transport in irradiated solids, (2) complex designs are used to keep these low thermal conductivity solids within their temperature limits under conditions of substantial nuclear heating and neutron damage, and (3) the resulting designs have a significant amount of non-breeding structure, coolant, and other materials. The primary safety uncertainties are the blanket tritium inventory, the tritium permeation rate into the coolant (water or helium) and the breeder tritium and thermal behaviour under transient conditions.

The R&D programs to date are attempting to resolve these and other issues. They range from generating property measurement data to testing of materials designed to achieve a significant burn-up in an advanced *in-situ* tritium recovery experiment. The range of experimental investigations includes material development and characterization, tritium recovery, study of breeder thermomechanical effects, examination of corrosion and mass transfer, and investigations of multiplier behaviour. These will lead to more complex tests with more relevant geometry, size and environmental conditions. Testing of non-nuclear submodules for thermomechanical properties are being carried out for design verification [41]. Nuclear submodule testing under irradiation to verify performance will also be a future requirement.

8. SUMMARY

Good progress has been made in the last five years on ceramic breeders. Irradiation testing has shown the feasibility of the proposed purge gas technique to recover the tritium produced in the ceramic. Progress is being made in understanding the mechanisms of tritium release so that the experimental results can be extrapolated to blanket operating conditions. Programs in material development, property measurements, and thermomechanical behaviour are also adding to the materials data base. Tritium recovery, tritium inventory and the material properties will all influence the selection of a ceramic material for a particular design. Much work remains to be done to clarify the issues, to be able to optimize the breeding material, the structural material and the neutron multiplier. Progress to date shows that these materials are viable for NET/ITER and future power reactors.

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