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TRITIUM BREEDING MATERIALS

G. W. Hollenberg, Westinghouse Hanford Company
C. E. Johnson, Argonne National Laboratory
M. Abdou, University of California at Los Angeles

ABSTRACT

Tritium breeding materials are essential to the operation
of D-T fusion facilities. Both of the present options -
solid ceramic breeding materials and liquid metal mater-
ials are reviewed with emphasis not only on their attrac-
tive features but also on critical materials issues which
must be resolved.

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I. INTRODUCTION

The on-site production of tritium at a fusion power plant is an established design practice for D-T fusion reactors; no other alternative is envisioned. Two classes of materials have evolved which can potentially satisfy the challenging engineering requirements of tritium producing blankets; liquid metals and solid ceramic breeder materials. Each of these two classes of material possesses its own special advantages and uncertainties when compared to the other. Although many other design options are available, no decision dominates the character of the blanket as much as the selection of the tritium breeding material.

After exploring the need for and history of tritium breeding materials, this review examines the major issues associated with the current classes of tritium breeding materials. A description of not only the current design concepts, but also the relevant data allows the reader to evaluate for himself the viability of these options. No attempt is made to prejudge the selection of tritium breeding materials, since our limited experience with these new (and sometimes exotic) materials provides a certain element of uncertainty in their application to the already complex fusion blanket.

II. THE NEED FOR BREEDING MATERIALS

The tritium breeding blanket provides three necessary functions in a fusion power plan, as shown in Figure 1.

- Acceptable tritium production (both breeding and recovery)
- Efficient heat recovery
- Acceptable neutron shielding

In addition, the blanket is required to be maintainable, safe, environmentally acceptable and possess an acceptable lifetime. Each tritium breeding material possesses specific assets in regard to fulfilling these functional requirements.

It is important to understand the absolute necessity of tritium producing blankets to the development of fusion reactors. First, the operation of D-T fusion plants will be difficult enough, so adopting even more challenging alternatives (D-D, etc.) does not appear appropriate for the first generation of fusion power plants. Secondly, the tritium appetite of D-T fusion power plants is clearly immense by present day standards. Unlike the vast supplies of deuterium in the oceans of the world, tritium for fusion plants must be a man-made substance.

As shown in Figure 2, a 3GWE device such as STARFIRE[1] would consume ∿0.5 Kg/day. All of the CANDU reactors in Canada[2] can provide about 0.008 Kg/day in the next few decades: a small fraction of the required tritium for STARFIRE. It is possible to produce tritium by irradiating lithium in fission reactors. If 10% of the neutrons in a 1GW fission reactor were used to produce tritium, over thirty fission reactors would be required to support one STARFIRE plant. Perhaps more efficient tritium production from fission reactors is possible, but an even more important point is that the adoption of fusion power by our society is predicated on the elimination of fission power by either resource or political limitations.

FIG. 1

FUNCTIONS OF A FUSION BLANKET

* TRITIUM PRODUCTION

* POWER PRODUCTION

INJECTION

T

D

CONVENTIONAL
THERMAL
POWER
GENERATION

PLASMA

TRITIUM
SEPARATION

TRITIUM
PRODUCTION

REACTOR
COOLANT

T INVENTORY

N N

N

FIRST WALL

SHIELD

MAGNETS

T RESERVES
AND DECAY

* NEUTRON SHIELDING

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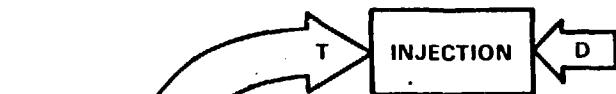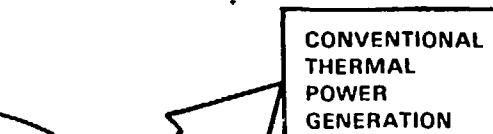
FIGURE 1.   The Three Major Functions of Fusion Tritium Breeding Materials are Tritium Production, Power Deposition and Neutron Shielding.

FIG. 2

THE NEED FOR A TRITIUM PRODUCING BLANKET

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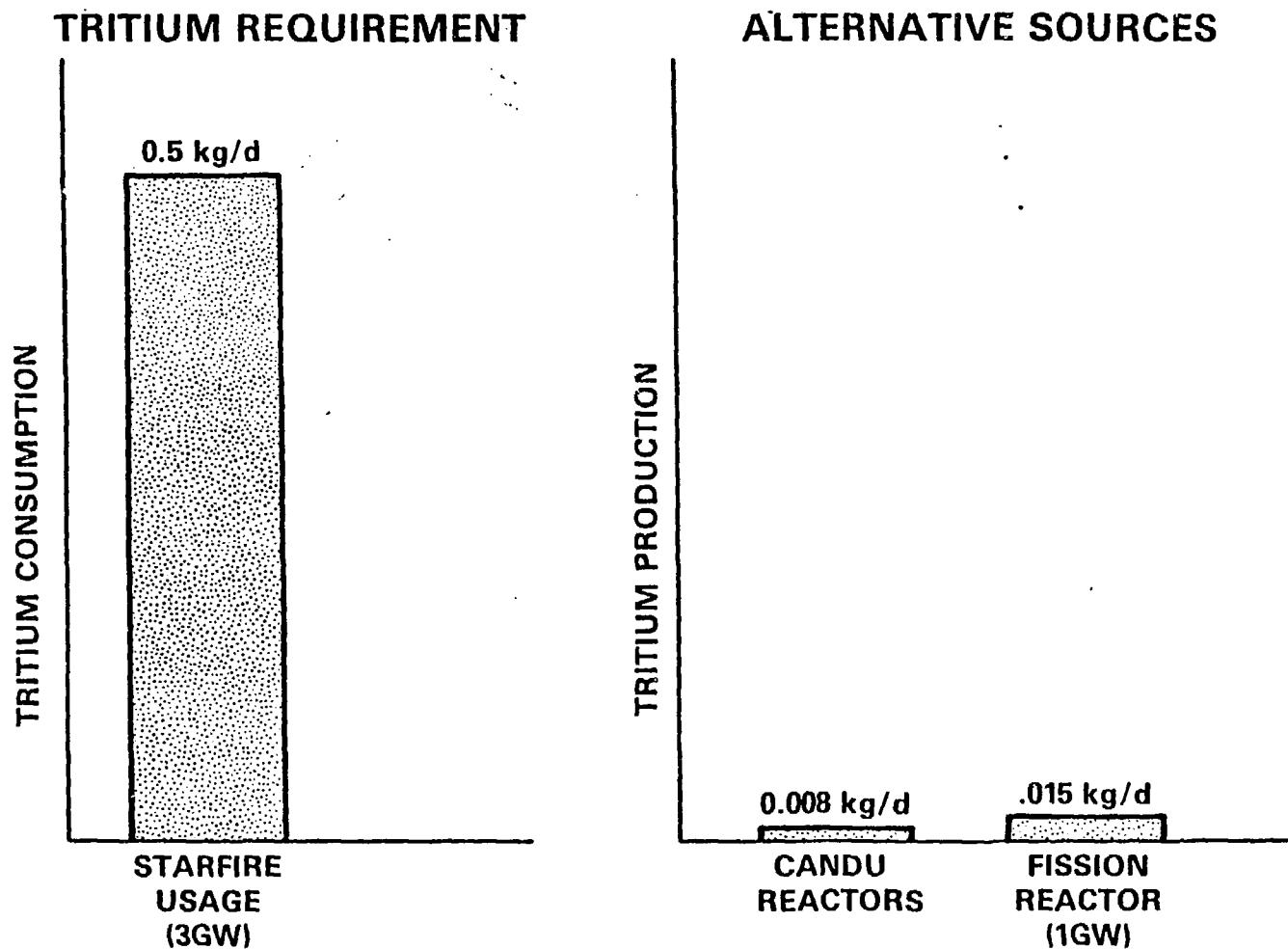
FIGURE 2. A Comparison of Tritium Requirements for a STARFIRE Fusion Plant from Sources Other than
Its Own Blanket Demonstrates the Necessity for High Tritium Production in the Blanket.

The use of lithium in tritium breeding blankets is based upon nuclear reactions with both ^{7}Li and ^{6}Li isotopes that actually produce more tritium in the blanket than is consumed in the plasma. The nuclear reactions with the two isotopes are distinctly different in that the ^{7}Li reaction, ^{7}Li(n,2n)^{3}T, is a threshold reaction with neutrons above 2 MeV, while the ^{6}Li reaction, ^{6}Li(n,a)^{3}T, possesses a "1/v cross section" which is highest at low neutron energies. There are two approaches to generating tritium: 1) pure lithium systems and 2) neutron multiplier systems.

In the pure lithium approach, shown in Figure 3, a high energy neutron from the plasma can react directly with a ^{7}Li atom or can be moderated until it reacts with a ^{6}Li atom. As shown in Figure 3, the ^{7}Li reaction is especially important because not only is a tritium formed directly but a secondary neutron from the reaction produces another tritium from a subsequent ^{6}Li reaction. However, the reaction of a moderated fusion neutron with a ^{6}Li produces only one tritium atom. Blankets with significant amounts of other substances, H_2O, Fe, Al, etc. moderate fusion neutrons so that the ^{6}Li single reaction dominates. Only breeder materials with high lithium atom density, like Li_2O and lithium metal, effectively use the ^{7}Li reaction which produces two tritium atoms from one fusion neutron.

In the case of neutron multiplier systems, designers take advantage of high-energy, neutron multiplication reactions that occur in several common isotopes, i.e., Be, Pb and Zr. For example, a single fusion neutron after reaction with beryllium produces two slower neutrons that can react with ^{6}Li atom to eventually produce two tritium atoms. In neutron multiplier systems it may be possible to use metals such as $Li_{17}Pb_{83}$ and low lithium atom density ceramics, i.e., $LiAlO_2$ and Li_2SiO_3. In reality, tritium breeding ratios, i.e., tritium atoms produced per fusion neutrons, when computed for the three dimensional blanket tend to be closer to 1.0 than 2.0. Recently it has been argued[3] that three dimensional breeding ratios (tritium created/tritium consumed) for the entire blanket of greater than 1.2 were consistent with a low risk design philosophy while a breeding ratio (BR) of 1.1 were indicative of a high risk design. The extra 10% or 20%

TRITIUM PRODUCTION REACTIONS

7Li REACTION CHAIN

FUSION NEUTRON E>2 MeV

7Li

3T

1n

4He

6Li

3T

4He

6Li REACTION

FUSION NEUTRON E<2 MeV

6Li

3T

4He
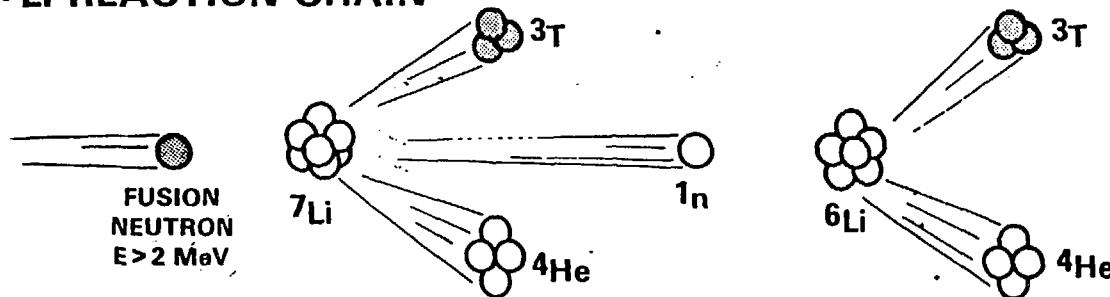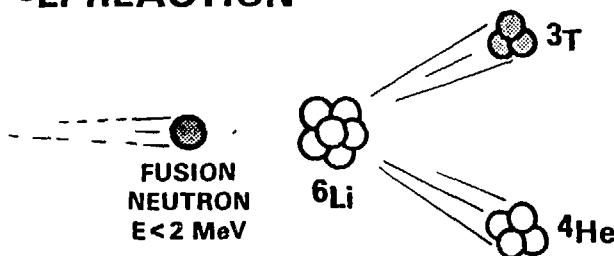
MULTIPLIER REACTION CHAIN

FUSION NEUTRON E>2 MeV

9Be

4He

1n

1n

4He

6Li

3T

4He

6Li

3T

4He

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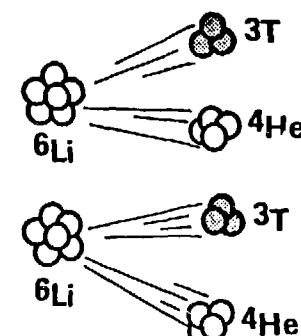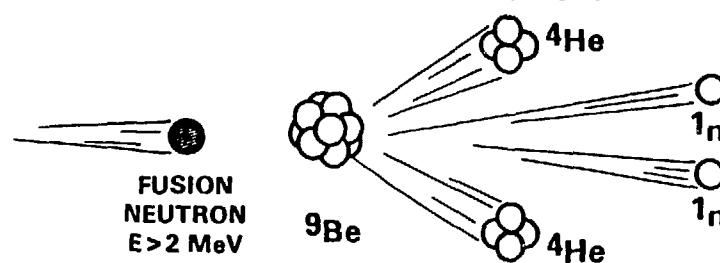
FIGURE 3. Pure Lithium Systems Produce Two Tritium Atoms When a High Energy Neutron Reacts With a ^{7}Li Atom, but Only One When Moderated Fusion Neutrons React Directly with ^{6}Li Atoms. Neutron multipliers, such as Be, can enhance tritium production in the blanket.

(i.e., BR ≈ 1.1 or 1.2) is necessary to allow for tritium decay, tritium inventory in the blanket, tritium inventory in other components, and start up supplies for future fusion reactors.

In many cases, there is an emphasis on the concentrated heat deposition on the surface of the first wall, but actually over 70% of the heat generated in a fusion plant is deposited in the tritium breeding materials. The necessity of the blanket to provide sensible heat extraction dictates in part the operating temperatures ($T > 250°C$) and configuration of the tritium breeding materials. Finally, the blanket provides much of the reduction of the neutron flux in front of the sensitive magnets.

III. HISTORY OF TRITIUM BREEDING MATERIALS

Deuterium was found to be separable by electroloysis of water in the 1930's, but tritium (with a concentration of only 10^{-18} in rain water) was more difficult to isolate and observe.[4]   Although Rutherford postulated the existence of tritium at this time, he first observed tritium by bombarding deuterons with deuterons in the Cambridge Accelerator. It was Alvarez[4] who first produced tritium in a manner we continue to follow today, i.e., exposure of lithium (metal) to a neutron flux.[4] In the 1940's, tritium was manufactured by irradiating LiF with subsequent thermal extraction.[5] In the 1960's, processes for extracting tritium from LiAl and $LiAlO_2$ were evaluated.[6-7] Hence, the production of tritium by lithium neutron capture significantly predates its adoption in blanket designs.

Fusion blanket design studies started to emerge in about 1972. Since then, blankets containing a number of materials: 1) for tritium breeding 2) structural material, 3) coolant, 4) neutron multiplier and 5) tritium recovery fluids were used as is shown in Table 1. The possible combinations of these materials together with a diversity of mechanical configurations have resulted in over 50 blanket concepts,[1,8-23] albeit with varying degrees of depth. Most of the emphasis has centered on liquid metals and solid ceramic breeding materials. While molten lithium salt's have been examined they were repeatedly dropped for a variety of reasons.

Since lithium, in some form, must be present in the blanket and since metallic lithium is an excellent heat-transfer fluid, the early generation (1972) design studies[8-11] considered self-cooled (circulating) liquid-lithium blankets. Solid breeding materials were considered in several studies for a variety of reasons. The earliest studies (1974) attempted to use solid breeding materials[1,19-23] to minimize lithium and tritium inventory by using a neutron multiplier and a lithium ceramic compound highly enriched in ^6Li. The neutron capture process was characteristic of the low energy neutrons used decades earlier for producing tritium in thermal fission reactors.

TABLE 1

CANDIDATE BLANKET MATERIALS

Breeding Materials	Coolants	Structure	Neutron Multiplier
Liquid Metals	Water	Austenitic Stainless Steel	Be
Li	(H_2O, D_2O)	Ferritic Steels	BeO
Li-Pb	Liquid Metals	Nickel-base Alloys	Pb
Li-Pb-Bi	Li	Refractory Alloys (e.g., V)	PbO
Molten Salts	Li-Pb		Bi
Flibe	Li-Pb-Bi		Zr
Intermetallic Compounds	Molten Salts		Zr_5Pb_3
Li_7Pb_2	Flibe		PbBi
Solid (Ceramic)	HTS		
Li_2O	Gases		
$LiAlO_2$	He		
Li_2SiO_3	Steam		
Li_2ZrO_3			
Li_2TiO_3			

Until about 1980, liquid-metal blankets were considered almost worldwide to be the leading option. In the course of the STARFIRE study,[1] the safety problems associated with liquid lithium received a great deal of attention and an advisory panel from electric utilities judged the safety problems as a serious objectional flaw in reactors using liquid lithium. Subsequent to the STARFIRE study, a significant R&D program on solid breeding materials was initiated as activities on liquid metals were reduced in the United States. Japanese R&D activities have had a singular emphasis on Li_2O throughout the 1970's and 1980's, while activities in Europe, until only recently, have soley emphasized liquid metals. Today, in programs such as the Blanket Comparison and Selection Study (BCSS)[24] a reassessment of critical issues is causing a reevaluation of both liquid metal concepts and solid breeder concepts.

IV. LIQUID-METAL BLANKETS

Liquid-metal blanket concepts can be classified into: 1) self-cooled, and 2) separately cooled. The most important liquid metals now considered are liquid lithium and $^{17}Li-^{83}Pb$. In the self-cooled concepts, the liquid metal is circulated as the coolant besides serving the functions of tritium breeding and neutron moderation.[24] In separately cooled concepts, another coolant is used for heat removal, while the liquid metal is only slowly circulated to recover tritium outside the reactor. Helium, water and molten salts are the candidates presently considered, but the use of water is ruled out ·in systems with liquid lithium.

One of the primary advantages of liquid-metal blankets is that salt extraction techniques for tritium recovery have been successfully demonstrated in laboratory-scale experiments that tritium concentration in liquid lithium can be kept to ∿1 kg under typical reactor conditions. The tritium inventory is predicted to be even lower for Li-Pb. A second advantage of liquid metal designs is that high tritium breeding ratios can be achieved, in fact Li-Pb systems have the potential of achieving breeding ratios greater than 1.5 with liquid lithium close behind.

From a safety standpoint, liquid lithium's strong chemical reactivity with air, water and even concrete continues to be a major design issue. There exists a considerable amount of operating experience available from LMFBR reactors for liquid sodium. But sodium is much less reactive than lithium as shown in the aftermath of the concrete reaction tests in Figure 4.[25] In comparison to sodium at 400°C, with a free energy of oxide formation with water of -50 Kcal/mole, lithium possesses a free energy of reaction with water of -140 Kcal/mole at 400°C, nearly three times as great. This highly exothermic reaction is part of the reason for the high reactivity and high temperatures (1300°C) generated in lithium/concrete tests. Hence, the use of liquid lithium necessitates, at a minimum, the use of protective measures such as multiple barriers to oxidation. The consideration of lithium-lead alloys is based on a much lower reactivity with air, water, and

CONCRETE/LIQUID METAL REACTIONS

LITHIUM
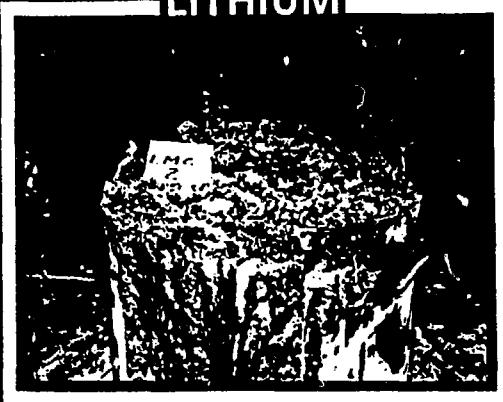
SODIUM
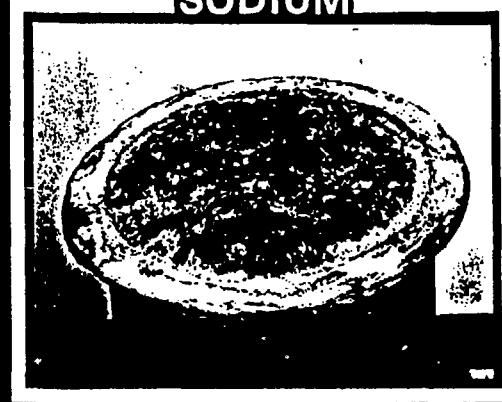
SELF HEATING TO 1300°C

END POINT:
MOLTEN STAINLESS STEEL
: LOSS OF LITHIUM
TO CATCH PAN

SELF HEATING TO 870°C

END POINT:
CONCRETE PENETRATION
TO 2.5 in.

FIGURE 4.   A Comparison of Reaction of Na or Li with Concrete at an Initial Temperature of 870°C.

concrete relative to lithium. The oxidation reaction of lead and water is actually endothermic (28 Kcal/mole) at 400°C. Of course, Li-Pb alloys have safety issues created by very low tritium solubility (~1 wppb). Low solubility produces high tritium partial pressures which induce tritium permeation through structural materials.

In the class of self-cooled liquid-metal blankets, ^{17}Li-^{83}Pb has been the most serious option considered[12,13] as an alternative to liquid lithium. On the negative side, Li-Pb is more corrosive to structural materials than liquid lithium. Corrosion can lead to thinning of the structural wall and transport redeposition of radioactive corrosion products in cooler areas of the system. In liquid-metal blankets, corrosion considerations dictate upper limits on the operating temperature at the interface between the liquid metal and structural material. Recent analysis suggests the temperature limits indicated in Table 2 based on a uniform dissolution rate of 5 μm/y imposed by mass transfer and redeposition considerations. As can be seen from Table 2, Li-Pb is more corrosive than lithium and would severely limit the use of austenitic stainless steel.

TABLE 2

CORROSION TEMPERATURE LIMITS* (°C)

| Liquid Metal | Flow | Structural Material | | |
		PCA	HT9	V-15Cr-5Ti
Lithium	Circulating**	430	535	750
	Static	525	565	750
17Li-83Pb	Circulating	375	415	650
	Static	395	435	650

*Based on Uniform dissolution rate of 5 μm/y (or ~5.5 mg/m^2 · h), see Reference 24.
**Velocity - 1.5 m/s.

The flow of liquid metals in the presence of the high magnetic field required for confinement results in induced currents and forces that oppose the fluid motion, normally called MHD effects. The resulting pressure drop can be unacceptably large in some self-cooled liquid-metal blanket concepts and remains a serious critical issue for the rest of the liquid-metal concepts. A large pressure drop requires operating the liquid-metal blankets at high pressure, negating one of the advantages thought for these systems in earlier generations of design studies.

The most serious issue is that MHD effects can impose on the structure stresses which exceed the allowable limits. Some designs attempt to reduce MHD pressure drops by: 1) flowing the liquid metal parallel to the main magnetic field wherever possible, and 2) reducing the fluid velocity by reducing ΔT_h through fluid mixing. To overcome the MHD problems with circulating liquid metals, blanket concepts in which the liquid metal is stagnant (in practice, slowly circulating for tritium recovery) with a separate coolant have been evaluated.[13,15] Another potential method for substantially reducing the MHD effects is to use insulators at the walls. However the viability of insulators in the irradiation field and other loading conditions in the fusion environment needs experimental investigation.

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V. SOLID BREEDER MATERIALS

Solid breeder materials can be classified into either BOT or BIT concepts. Breeder out-of-tube (BOT) concepts such as STARFIRE[1] feature a porous ceramic solid enclosed by external structural materials, such as stainless steel, with coolant tubes and helium purge gas channels for insitu recovery of tritium. Breeder-in-tube (BIT) concepts such as proposed in BCSS[24] contain the solid breeder in tubes, much like fission reactor fuel, but then a large structural enclosure must contain the coolant, i.e., helium.

In lieu of operating data on solid breeder materials in actual fusion reactors, it was necessary to theoretically predict the performance of these materials. Recent contributions from laboratory experiments and fission reactor irradiation have significantly improved the data base for using these materials. The inherent refractoriness and stability of the ceramic tritium breeding materials provides the incentive for their further development but several critical issues are yet to be fully resolved; i.e. tritium inventory, lithium transport and mechanical interaction.

In Table 3, the lithium atom density of Li_2O is contrasted with that of the ternary oxides.[25] Only Li_8ZrO_6, a relatively new material, even approaches the lithium density of Li_2O. In fact, Li_2O has a higher lithium density than even lithium metal (0.53 gm/cm^3). $LiAlO_2$, in contrast, possesses only a third of the lithium atom density of Li_2O which is reflected in Figure 5 as a lower breeding ratio. The high lithium atom density in Li_2O allows it to be incorporated into designs which can result in breeding ratios of greater than 1.5, while $LiAlO_2$, even with a neutron multiplier, obtains a "low risk" breeding ratio, 1.3.[2] The continued interest in $LiAlO_2$ is maintained by its wide acceptable temperature operating range in comparison to Li_2O. If a narrower operating temperature range for Li_2O is identified in the future, a blanket design based on Li_2O would prove to be difficult to operate.

FIG 5

SOLID BREEDER MATERIAL CANDIDATES

RANGE OF POTENTIAL BREEDING RATIOS

Li_2O
WITH MULTIPLIER

$LiAlO_2$-
WITH MULTIPLIER

Li_2O
NO MULTIPLIER

$LiAlO_2$
NO MULTIPLIER

1.5

1.4

1.3

1.2

1.1

1.0

0.9

0.8

300   500   700   900   1100   1300

RANGE OF ACCEPTABLE TEMPERATURES
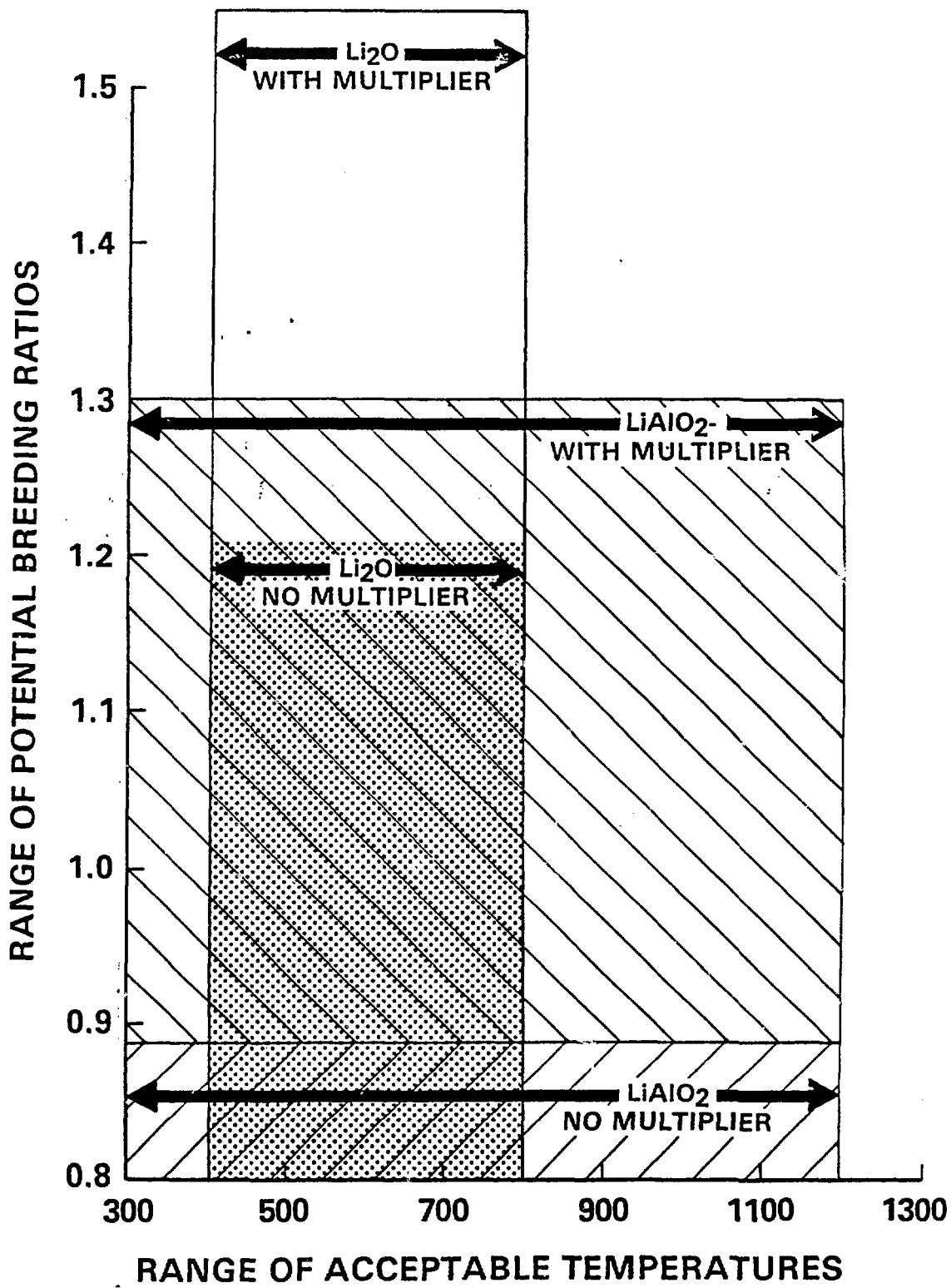
FIGURE 5.   The High Tritium Breeder Ratios of Li_2O are Contrasted with Its
            Small Operating Temperature Range in Comparison to $LiAlO_2$.

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TABLE 3

PROPERTIES AND RECOMMENDED TEMPERATURE LIMITS
FOR CANDIDATE SOLID BREEDER MATERIALS

	Properties			Recommended Temperature Limits		
	MP (°C)	ρ_{Li} (g/cm^3)	K (W/MK)	T_{min} (°C)	T_{max} (°C)	ΔT (°C)
Li_2O	1433	0.93	3.4	410	800	390
Li_8ZrO_6	1295	0.68	1.5	350	980	630
γ-$LiAlO_2$	1610	0.28	2.2	300	1200	900
Li_5AlO_4	1047	0.61	2.3	350	780	430
Li_2SiO_3	1200	0.36	1.5	410	1000	590
Li_4SiO_4	1250	0.54	1.5	320	950	630
Li_2ZrO_3	1616	0.33	1.3	400	1400	1000

The tritium inventory of an operating solid breeder blanket may be its single most important performance parameter and is essentially intrinsic to a specific material under specific conditions. The tritium inventory is thought to be partitioned into several distinct phenomena; solubility, bulk diffusion, irradiation effects and gas diffusion within the pores. While selection of an inventory limit - be it 10 wppm or 1% - is controversial, candidate materials exhibiting low inventory are clearly more desirable. The tritium inventory causes an extra burden on breeding ratios for solid breeder designs. The tritium inventory within a candidate breeder materal is predicted to possess a strong dependence on temperature and an uncertain dependence on burnup.

The solubility contribution to the tritium inventory was originally thought to be the dominant factor. Currently, only Li_2O has received considerable attention in laboratory studies.[27] Even though hygroscopic at room temperature, Li_2O has been shown to have extremely low moisture solubility under blanket operating conditions as shown in Figure 6. However,

LiOT SOLUBILITY IN Li₂O

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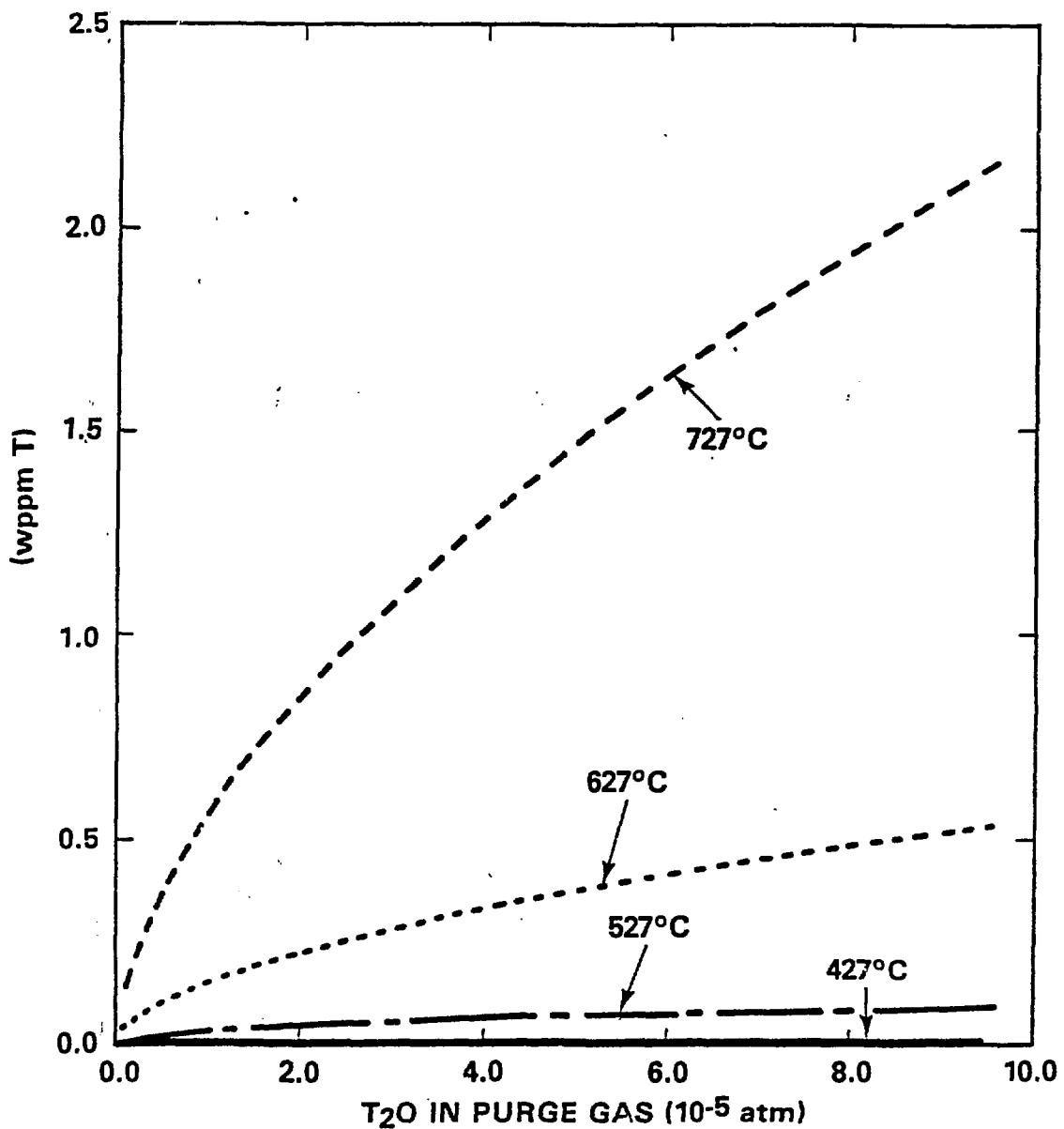
FIGURE 6.   Data on the Solubility of LiOT in Li_2O Indicates Much Lower
            Solubility of LiOT in Li_2O than First Predicted.

if the T_2O partial pressure were to increase in the gas phase over the sample, a separate LiOT phase would form with an incremental increase in tritium inventory.

These laboratory studies were complimented by recent in situ recovery experiments on Li_2O in a fission reactor in Japan[28] which indicated only 0.5 wppm tritium retained at 750°C. In addition, the recent TRIO-01 experiment in ORR[29] demonstrated in situ tritium recovery from γ-$LiAlO_2$ while under fission neutron irradiation. These experiments were conducted in part with He-H_2 purge streams which appeared to significantly enhance tritium release in comparison to less reducing atmospheres. The chemical form of tritium being released, i.e., condensible (T_2O,HTO) or noncondensible (HT,T_2), continues to be a topic of interest and conjecture since it directly impacts containment and processing systems.

Irradiation effects, including restructuring, trapping and defect generation, can also contribute to the tritium inventory.[30] Although the degree of irradiation induced trapping is still uncertain, fast reactor closed-capsule irradiation experiments have served to demonstrate that irradiation effects will be limited to a few percent (1 to 9%) of the tritium generated in Li_2O.[30] Restructuring, in particular grain growth, was once thought to provide an upper temperature limit, but data now indicate that the effect of temperature on diffusion is more dominant than any decrease resulting from larger grain sizes. Interestingly, $LiAlO_2$ and Li_4SiO_4 retained more tritium in this fast reactor environment and at lower temperatures which indicates mechanisms other than solubility to be rate controlling, for example, surface or diffusional phenomena.

In Table 3, the upper temperature limit for Li_2O results from concerns over gas phase transport of LiOT which would relocate blanket material from hot regions into cold regions which are in the path of tritium leaving the blanket. In Figure 7, the vapor pressure of LiOT over Li_2O, with a 10^{-5} atm T_2O pressure, increases dramatically at approximately 800°C.[31] Whereas, temperatures of over 1000°C are required for lithium or lithium-oxide gas

MASS TRANSPORT IN Li$_2$O
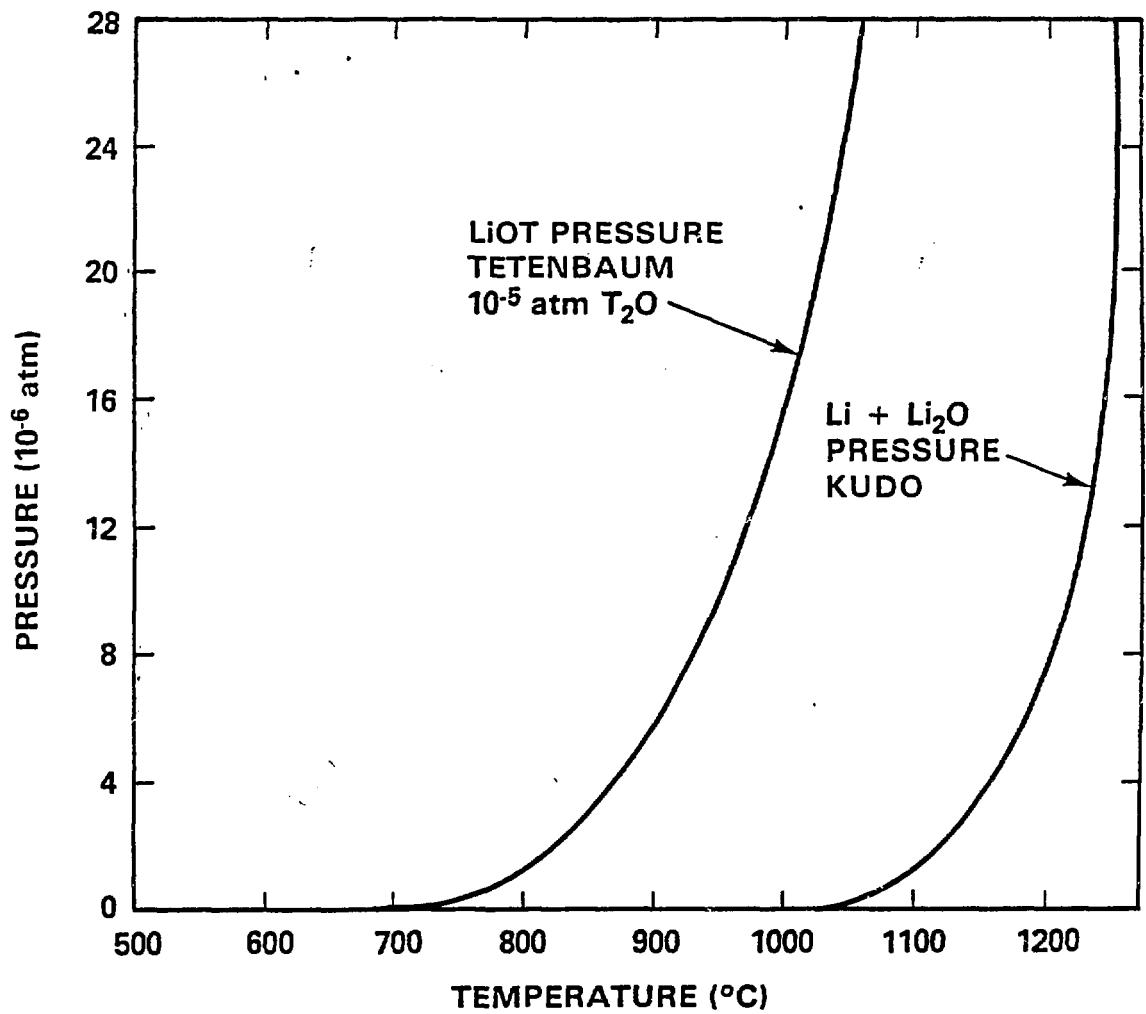

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FIGURE 7. Gas Phase Mass Transport in Li_2O is Controlled by the LiOT Vapor
Pressure Which is Very Temperature Dependent.

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species to reach significant vapor pressures.[32] In actual fast reactor irradiation experiments on Li_2O, at 900°C over 0.5% of the lithium in a pellet was transferred to cold capsule walls.[30] Again, there is continued controversy on the true significance of LiOT transport. In designs that feature temperatures above 800°C LiOT transport is thought to be localized by directing purge gas flow in only cold regions (T > 700°C).

Recently, swelling of fast neutron irradiated Li_2O at 500°C to 900°C has been observed,[30] but little swelling is evident in ternary oxides ($LiAlO_2$, Li_4SiO_4 and Li_2ZrO_3). Swelling in Li_2O is thought to be the result of helium retention in bubbles within individual crystallites. The swelling in Li_2O was found to be 1.2% to 1.8% at a burnup equivalent to roughly half a year of operation in a 5 MW/m^2 wall loading fusion plant. The potential impact of unrestrainable swelling on the operation of a fusion power plant is that mechanical interaction between the solid breeder and structural components will lead to a loss of mechanical integrity of the blanket and premature blanket replacement. Fortunately, Li_2O pellets can be hot pressed at only 700°C, consequently, dilatational strains may be adequate in certain configurations to accomodate swelling.

VI. CONCLUSIONS

An indisputable need of D-T fusion power remains the definition of a reliable, safe and functional material which efficiently breeds tritium. Although two material classes - solid and liquid breeder material - have been used as the basis for reasonable blanket concepts, critical issues and uncertainty still remain for both.

Liquid lithium is mainly fraught with safety (reactivity) concerns, but MHD effects may also be of consequence. Although lithium lead alloys are not as limited by reactions with oxygen, the corrosion of structural materials by $^{17}Li^{83}Pb$ and tritium mobility forcasts a separte class of risks.

The inherent chemical stability of ceramic oxides stimulates the search for solid breeder materials unencumbered by these safety issues. But the accessibility of tritium generated in these materials and their ability to adequately generate tritium are questions which require early answers. In addition, lifetime limiting issues such as lithium transport and swelling, may decide the fate of the otherwise favored Li_2O, or may not enter in at all.

Our immediate concern is that tritium breeding materials' issues must be resolved, allowing us to be poised with viable blanket options of reasonable risk. But in the long run, the investment in fusion power plants unavoidably dictates the verification of every aspect of tritium breeding materials operation, i.e., safety, tritium recovery, lifetime, etc., to the same degree as any other component in the plant.

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