THE DEVELOPMENT OF TRITIUM BREEDING BLANKETS

FOR

DT-BURNING FUSION REACTORS

MASTER

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THE DEVELOPMENT OF TRITIUM BREEDING BLANKETS

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Summary

This study examines the status of under-
standing of blanket tritium recovery and the
performance of potentially viable tritium
breeding materials under conditions anticipated
in a DT-fueled fusion reactor environment. The
existing physicochemical, thermophysical, and
ceramographic data for candidate liquid and solid
breeders are reviewed and appropriate operating
conditions defined. It is shown that selection
of a breeding material and an approprinte tritium
recovery method can impose significant con-
straints upon blanket design, particularly when
considerations of breeder/coolant/structure
compatibility and temperature limitations are
taken into account.

Introduction

Lithium appears to be the only element
suitable for breeding tritium in a commercial
tokamak reactor. Of the large number of lith-
ium-bearing materials, only a few types of ma-
terials appear to be the most promising for
tritium breeding. These include liquids, such
as elemental lithium itself and certain alloys
(e.g., $Li_{0.17}Pb_{0.83}$); and solids, such as inter-
metallic compounds (e.g., Li_7Pb_2), lithium oxide
(Li_2O), and ternary oxides (e.g., α-$LiAlO_2$,
Li_2ZrO_3, etc.).

For each candidate breeder, a number
of performance requirements can be used as a
measure of the relative merits of each candidate
breeding material, namely:

1. tritium breeding,

2. chemical and physical stability,

3. tritium recovery,

4. compatibility with coolant,

5. compatibility with structure.

6. thermophysical properties,

7. economics, and

8. safety.

In the context of this study, a comparative eco-
nomic analysis is not possible, owing to the
rather limited data base. Also, safety issues
will be discussed in qualitative terms. The
focus of this study is primarily upon tritium
recovery and appropriate operating conditions for
each breeding material. The anticipated per-
formance of each material under commercial tokamak
reactor conditions, which include radiation,
temperatures, magnetic fields, and effects of
lithium burnup and helium generation is reviewed.

Breeding Materials

The subject of liquid metal breeding materials
in tokamak reactors has been recently reviewed[1]
and was discussed as part of a comprehensive
blanket study.[2] There is another liquid that could
be considered, the molten salt, LiF-BeF_2 ("Flibe")
but it is not capable of attaining adequate tritium
breeding,[3] and it is, therefore, not considered in
this study. Three liquids appear to be the most
attractive, namely, elemental lithium itself, the
eutectic $Li_{.17}Pb_{.83}$, and the ternary mixture
$Li_{.1}Bi_{.5}Pb_{.4}$. The data base for lithium is ex-
tensive and most of the physical and chemical
properties of primary importance, e.g., Sieverts'
constants are known.[4,5] Owing in part to the ex-
tensive data base, lithium was selected as the
breeding material for many design studies.[6-13]
The compound $Li_{.17}Pb_{.83}$ (considered as a breeder
for the INTOR Study[14]) is much less well-char-
acterized, but the Li-Pb phase diagram has been
known for nearly 50 years and the melting point
of $Li_{.17}Pb_{.83}$ is 235°C (cf. 180°C for elemental
lithium).[15] The density of various Li-Pb compo-
sitions was measured[16] and the density of liquid
$Li_{.17}Pb_{.83}$ is estimated to be 9.4 g/cm³ at 235°C.
Lithium activities have been measured[17] and
found to be about 10^{-4}, four orders of magnitude
below those of lithium. In a preliminary experi-
ment, the Sieverts' constant for dissolution of
D_2 in $Li_{.17}Pb_{.83}$ at 770°C was measured to be about
200 atm$^{1/2}$/mole fraction D,[17] two orders of magnitude
higher than that of liquid lithium. The quali-
tative reactivity of various lithium-lead materials
was tested at Argonne National Laboratory by heat-
ing samples to 500°C and dropping them into
water.[18] It was found that $Li_{.17}Pb_{.83}$, in con-
trast to liquid lithium and lithium-rich alloys,

has only very modest reactions with water.[18] Thus, this alloy does appear to offer a significant safety advantage. The use of Li-Pb alloys as breeders for fusion reactions is discussed in another study.[19] Ternary Li-Pb-Bi systems have not been previously reported, but in a series of recent experiments at Argonne National Laboratory the melting point of $Li_{.1}Pb_{.5}Bi_{.4}$ was found to be 140°C.[20] Although there is essentially no data for this ternary alloy, it is expected to be similar to $Li_{.17}Pb_{.83}$.

Tritium Recovery

The feasibility of recovering tritium from liquid lithium has been demonstrated and it appears that tritium levels of ≤ 2.0 appm (≤ 1 wppm) are achievable by molten-salt extraction.[21] This would translate to a blanket-tritium inventory of ~ 100 g per GW of fusion power. However, it must be noted that demonstration of the full-scale engineering feasibility of the method for continuous tritium recovery remains to be achieved. Nonetheless, molten salt extraction also should be a suitable method for recovering tritium from $Li_{.17}Pb_{.83}$ and $Li_{.1}Ph_{.4}Bi_{.5}$. In a preliminary experiment the solubility of Pb in the lithium halide salt used for extraction was found to be less than about 10 wppm.[22] Therefore, mutual solubilities of salt and alloy are expected to be acceptably small. Also, in contrast to Li (density 0.5 g/cm³) the density of the alloys (~ 10 g/cm³) is much greater than that of the salt (2.6 g/cm³). Thus, for the alloys, the extraction process is simplified and in the event of carry-over of the liquid alloy into the salt, the alloy will not interfere with the electrolysis step used to recover the tritium (LiT) from the salt.[22]

From thermodynamic considerations, because the Sieverts' constants for the alloys are about two orders of magnitude greater than those of lithium, the attainable tritium inventories in the alloys would be about two orders of magnitude lower than in liquid lithium, i.e., ~ 1 g/GWth. However, mass-transfer considerations would make it most difficult to attain tritium levels that low. The inventories still could be lower than those in liquid lithium, probably the order of ~ 10 g/GWth. This means that liquid alloys are expected to have the lowest tritium inventories of any system.

On the other hand, because the Sieverts' constants are high for the alloys, tritium pressures will be relatively large, about two orders of magnitude higher than for lithium itself. As a result, tritium permeation rates will be a significant concern. In other words the tritium inventory in the alloys will be very small, but the tritium will be relatively mobile.

Anticipated Performance of Liquid Breeders

The properties of the three liquid breeders considered are summarized in Table 1. All three appear to have excellent tritium breeding capability and the prospects for recovery of tritium from these materials are very good. It is likely that tritium inventories will be very low. Lithium is reactive with water, but it appears that proper design can prevent the accidental contact of lithium with water.[1] Lithium should not be used with water coolant, but it can be used with either helium or lithium as the coolant.[1] Because the liquid alloys are much less reactive with water, it is conceivable that they could be used with any coolant, including water. In Table 1, no upper temperature limit is given. That is because that limit depends upon the maximum temperature of the structural material, thus it is strongly design-dependent.

Breeder/structure compatibility and corrosion is a major concern with liquid metal breeders. There is a reasonable data base for the corrosion of alloys by liquid lithium. Lithium is generally compatible with refractory metals, but it is not compatible with nickel-base alloys.[2] For titanium alloys and ferritic steels, corrosion and mass transfer limits the operating temperature to a maximum of ~ 500°C.[2] The maximum temperature is lower for austenitic stainless steels.[2] Corrosion of structural materials by the liquid alloys is not as well understood at this time, but it is a matter of significant concern. This topic is discussed in another paper.[19]

Radiation effects upon liquid breeders should present no problems at all. Lithium will have to be replaced to replace that which is burned up, but that is easy to do. Significant amounts of helium gas will be generated (a few standard cubic meters per day in a commercial tokamak reactor), but it appears that appropriate design solutions exist.

The impact of magnetic fields can be significant, particularly if the liquid metal is used also as a coolant. Although MHD effects are a matter of concern, it appears that pumping power can be reasonable (< 1% of the thermal power), and hydraulic pressures are comparable to, or less than, those of other coolants.[1]

Solid Breeders

The design implications of using solid breeders was recently reviewed[23] and also these materials were extensively studied in the STARFIRE design.[24] In addition, two related papers are being given at this meeting, one presenting the analyses of tritium release mechanisms[25] and one discussing materials selection[26] for STARFIRE. Solid breeders offer a number of potential safety advantages, including relatively low tritium mobility and low stored chemical energy. Potentially viable solid breeders include two major types, namely, (1) alloys and intermetallic compounds, and (2) oxides. Examples of intermetallic compounds are Li_7Pb_2, LiAl, and Li_3Bi. The eutectic $Li_{.62}Pb_{.38}$ has been proposed[27] as a

Table 1. Summary: Liquid Metal Breeders

Composition	m.p.,°C	Tritium Breeding	Tritium Inventory, g/GWth	Reactivity With Water	Operating Temp. Range, °C
Li	180	Excellent	~ 100	Vigorous	230-*[c]
$Li_{.17}Pb_{.83}$	235	Excellent[a]	~ 10	Mild	300-*[c]
$Li_{.1}Pb_{.4}Bi_{.5}$[b]	140	Excellent[a]	~ 10	Mild	200-*[c]

[a] Pb (also Bi) is a neutron multiplier. Enrichment of ^6Li is required.

[b] Resources of Bi are limited.

[c] Maximum temperature is set by structural material.

two-phase breeder. The oxides include Li_2O and certain ternary oxides, e.g., $LiAlO_2$ and Li_2ZrO_3.

Tritium Recovery from Solid Breeders

Tritium recovery from solid tritium-breeding materials was identified as a key factor in establishing the viability of the solid-breeder concept and an important consideration in the selection of the primary candidate materials for the STARFIRE design. In-situ tritium recovery from fusion blankets is by far the most desirable if not the only viable method of tritium recovery. Mobile solid blanket concepts have been proposed[28-30] as a means of tritium recovery, however, the feasibility of this concept is questioned primarily on the basis of breeding capability and design complexity, particularly for the tokamak configuration. The economics of a batch-type tritium recovery scenario is generally regarded as unacceptable for a power reactor system.[2]

In the STARFIRE design study, tritium was removed from the blanket by means of a purge stream, which flowed through formed purge channels in the blanket. Five specific mechanistic steps required for bred tritium to be recovered were identified, namely:

1. bulk diffusion,

2. desorption of tritium (T_2O),

3. grain boundary migration,

4. "percolation" of tritium through the pores in the solid to the purge channel, and

5. convective mass transfer out of the blanket via the purge channels.

Diffusion of Tritium in Solids

The tritium recovery experiments that have been performed on solid breeders[31-36] are limited to short-term, low-temperature irradiations with post-irradiation tritium recovery. Therefore, tritium concentrations generated in these experiments were low, the neutron fluences were low, and the radiation temperatures were below those of interest. The data from these experiments represent a combination of several of the five steps, primarily the first three and part of the fourth step. In an effort to better understand the processes involved in tritium recovery, a number of models have been developed.[25]

Before discussing the results of the models, a further set of definitions is useful. There are two ideal limiting cases for determination of the tritium inventory. First, one may assume that the tritium inventory is diffusion controlled (i.e., step 1 predominates). Accordingly, Ficks' Laws are solved assuming steady state, a geometry (e.g., spheres), and boundary conditions (e.g., zero surface concentration). The diffusional model gives a tritium inventory resulting from the concentration gradients within individual grains. Since this model usually assumes that the tritium concentration at the grain edge is zero, the calculated tritium inventory is obviously lower than the true inventory.

Another approach is to assume chemical equilibrium, with a fixed tritium (or T_2O) pressure in the gas phase in equilibrium with the solid. This method assumes uniform concentrations (activities) of tritium throughout the blanket. Because this model does not account for concentration gradients, tritium inventories are again underestimated. For this analysis, the tritium inventory is assumed to be the sum of a diffusive component and an equilibrium component. The total inventory is then approximated as the sum of the diffusive inventory and the solubility inventory.

Bulk diffusion is considered to be a significant contributor to the tritium inventory. For spherical particles of radius r, assuming zero surface concentration, the tritium inventory I is given by

$$I = \frac{1}{15} \dot{I} \, r^2/D \qquad (1)$$

where $\dot{I}$ = the tritium generation rate and D = diffusivity. It is very significant that the tritium inventory is a function of the square of the particle size. Clearly, small ($\sim 1\,\mu$) grain sizes are required for minimum tritium inventory and the grains must not enlarge significantly during the lifetime of a reactor blanket. While diffusivity values for hydrogen have been measured for many solids, there is a rather large uncertainty for what the correct values for tritium in solid breeders will be. Kinetic experiments of post-irradiation tritium release from several candidate solid breeders have been performed. In these experiments, the kinetics are non-steady state and the diffusivity D is given by:

$$D = 0.16 \, r^2/\tau \qquad (2)$$

where τ is the mean residence time defined as the time required to extract 87.4% of the tritium.[34]

Because the grains in experiments are of an very small ($\sim 1\,\mu$), non-spherical, and non-uniform, it is very difficult to determine r and, therefore, D in Eq. (2). However, Eqs. (1) and (2) can be combined yielding:

$$I = 0.5 \, \dot{I} \, \tau \qquad (3)$$

Since τ is readily measured, one can estimate the diffusive inventory in a blanket, provided the particle microstructure in the blanket corresponds to that in the experiments. The tritium generation rate per GWth assuming a breeding ratio of 1.2 and 20 MeV/fusion is 1.87×10^{-3} g/s. Substitution into Eq. (3) yields

$$I = 3.3 \cdot \tau(h) \qquad (4)$$

where I = the diffusive tritium inventory in grams per GWth and $\tau(h)$ = the mean residence time determined in post-irradiation annealing experiments. Accordingly, the data of Wiswall[31] were used to obtain the results in Fig. 1.

The results of the above somewhat empirical model show a temperature dependence which is consistent with a diffusion-controlled process. The model is based on diffusion rates and makes the assumption that grain size and geometry in the blanket are the same as those in the experiments. It is interesting to compare Fig. 1 with a figure compiled by Sze,[37] which shows a very wide scatter in reported diffusivity values of tritium in various candidate solid breeders. One reason for this apparent discrepancy is that
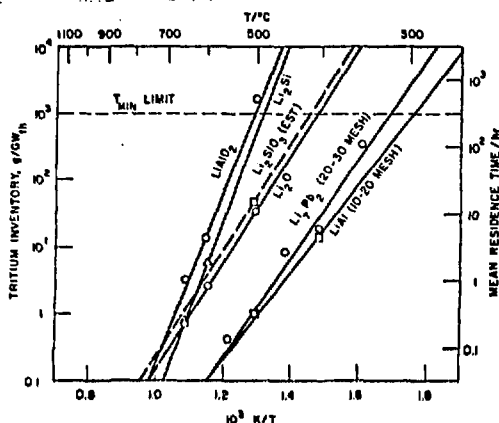

Fig. 1. Release of tritium from solid breeders

the diffusivity is a function of the square of the diffusion path length r (e.g., Eq. (2) above) and for a solid having various grain sizes and shapes it is very difficult to determine r. Thus, although diffusivity values are not well known, diffusion rates are apparently more consistent. It is also noted that an independent approach,[28] using well-known diffusivity values for hydrogen in single-crystal Al_2O_3, were in good agreement with the values for $LiAlO_2$ in Fig. 1.

Restructuring or grain growth could greatly affect the diffusive inventories since the calculated inventory varies as the square of the grain size. The sintering characteristics of the candidate breeding materials have not been investigated. In general, the thermal sintering characteristics of several stable oxides are similar. Temperatures in excess of 0.8 T_m (the absolute melting point) are required before significant sintering occures. However, neutron radiation typically enhances the sintering characteristics and lowers the temperatures at which sintering is observed. The effects of radiation are expected to reduce the sintering temperature to 0.6 T_m.[24,25]

Based on the present model, which combines aspects of diffusion theory with experimental data, the range of operating limitations are defined where bulk diffusion is the rate-limiting step. A minimum temperature is defined as the temperature at which the tritium inventory exceeds 1 kg/GWth (Fig. 1). Since, there will be a range of temperatures in the blanket, the inventory distribution must be determined by integrating the tritium generation rate and the temperature distribution throughout the blanket. In the present analysis the meaning of "T_{MIN}" in Fig. 1 assumes that no more than 5% of the blanket can be at temperatures below T_{MIN} and none of the blanket can be more than 50° below T_{MIN}. Based upon the above criteria, the minimum temperatures for

solid breeders predicted from diffusion controlled processes are given in Table 2.
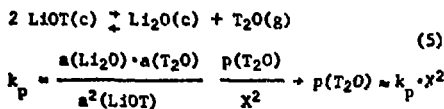
Table 2. Temperature Limits (°C) for Solid Breeders

Breeder	$T_{min}/°C$	$T^*_{max}/°C$
Li_2O	410	910
$LiAlO_2$	500	850
Li_2SiO_3	420	610
Li_2TiO_3	420	820
Li_2ZrO_3	420	860
Li_2Si	480	350
$LiAl$	300	310
Li_7Pb_2	320	330
Li_3Bi	320	580

*
0.6 absolute M.P.

The resulting operating temperature limits (Table 2) have significant design implications. For example, with the exception of Li_3Bi, the intermetallic compounds appear to have unacceptable operating temperature ranges and these materials do not appear attractive for in-situ tritium recovery.

Thermodynamic Equilibria and Tritium Dissolution

The tritium inventories in the candidate solid tritium-breeding materials that result from the thermodynamic solubility have also been evaluated. The tritium concentration in the grains are predicted as a function of the tritium or T_2O partial pressures in the gas phase. Since no data have been published on the solubility of tritium in the candidate breeding materials, a thermochemical model was used to estimate the solubilities in the oxides. The assumptions used in the model include: (1) JANAF thermochemical data[38] for hydrogen were used, (2) isotope effects were not considered, (3) tritium was assumed to be in the form of LiOT in solid solution in the breeder, and (4) activity coefficients of all species were assumed to be unity. Assumptions 3 and 4 combined may be considered as Raoult's Law. For a specific example, consider Li_2O:

$$2 \text{ LiOT(c)} \rightleftarrows Li_2O(c) + T_2O(g)$$

$$k_p = \frac{a(Li_2O) \cdot a(T_2O)}{a^2(LiOT)} \frac{p(T_2O)}{X^2} \rightarrow p(T_2O) \approx k_p \cdot X^2 \qquad (5)$$

where X is the mole fraction of LiOT in Li_2O.

For ternary oxides, the relevant equilibrium reaction is expressed as:

$$2 \text{ LiOT} + MO_x \rightleftarrows Li_2MO_{x+1} \qquad (6)$$
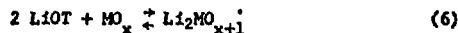
and $p(T_2O) = X_M \cdot X^2 \cdot K_p$

In the above expressions, $p(T_2O)$ is the partial pressure of T_2O, X is the mole fraction of LiOT, and X_M is the mole fraction of the binary metal oxide (e.g., Al_2O_3). For the systems of interest, the activities of LiOT and MO_x have not been experimentally determined. The activity of LiOT is probably within a factor of ten of the mole fraction if LiOT is in solution. However, there is a greater uncertainty for the activity of the lithium depleted species, e.g., Al_2O_3, for two reasons:

1. The lithium depleted species may not be a simple metal oxide (Al_2O_3) but most probably is another ternary compound such as $LiAl_5O_8$. Since $LiAl_5O_8$ forms, it is more stable than Al_2O_3, and therefore, free energy changes and k_p values for for reactions such as (6) may be overestimated.

2. The composition of the breeding material continuously changes during operation of the blanket because the lithium is being burned up. Since tritium is being removed from the blanket, the mole fraction of LiOT will reach a constant value. However, the concentration of the other specie, e.g., Al_2O_3, will continuously increase at a rate of about 1% per year.

Although there is considerable uncertainty in the value of X_M and because it changes with time, a conservative value of $X_M = 0.5$ X is assumed for the preliminary scoping studies. Using the above expressions, equilibrium tritium concentrations in solid ceramic breeders for a T_2O partial pressure of 1.3 Pa (10^{-2} torr) were calculated (Fig. 2). It is evident that tritium dissolution in Li_2O appears to be unacceptably high for the temperatures of interest. All of the ternary oxides appear to have significantly lower tritium "solubilities" and, with the possible exception of Li_2ZrO_3, have levels of ~ 10 wppm or less. Experimental data are needed to verify the predictions of the model.

Anticipated Performance of Solid Breeders

The physical properties, tritium recovery characteristics and neutronics properties are summarized in Table 3. Of the intermetallic compounds, only Li_3Bi appears to have a workable operating temperature range. However, Bi is resource-limited and it is questionable that there is sufficient abundance to use Li_3Bi as the breeder for commercial fusion reactors. In addition, Li_7Pb_2 and
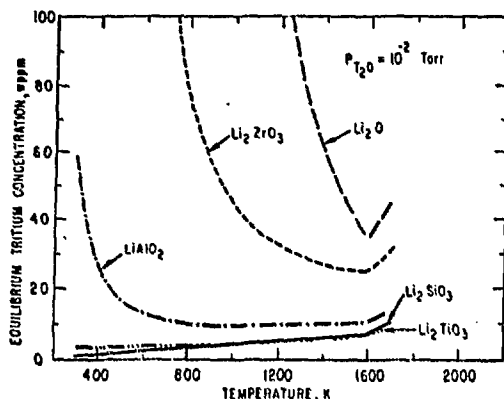
Fig. 2. Calculated T_2O solubility in various solid breeders.

Li $_{62}$Pb $_{38}$ were shown to react vigorously with water.[18] Therefore, the listed alloys and intermetallic compounds do not appear to offer a significant safety advantage compared to liquid lithium. Thus, for the above reasons, the intermetallic compounds do not appear to be attractive. The oxides, owing to their chemical stability and high melting points, appear to be the most promising solid breeder materials.

Thermochemical calculations indicated that lithium oxide has very high tritium solubility, to the extent that tritium inventories in Li_2O may be unacceptably high. The ternary oxides appear to have lower tritium solubilities, and thus are likely to have lower tritium inventories. The ternary oxides, however, require a neutron multiplier. It is noted that the STARFIRE study identified two multipliers (Be, Zr$_5$Pb$_3$) which appear to be suitable.[24]

A major concern regarding the use of solid breeders is radiation effects.[24,25] Continuous, in-situ recovery from solids requires that the

Table 3. Solid Breeders

	m.p. °C	Temperature Range, °C	Tritium Dissolution	Li Atom Density g/cm^3	Breeding Capability
Intermetallic Compounds and Alloys					
Li$_7$Pb$_2$	726	320-330	Low	0.49	Excellent
Li$_3$Bi	1145	320-580	Low	0.47	Excellent
LiAl	700	300-310	Low	0.36	Fair[b]
Li$_2$Si	760	480-350	Low	0.36	Fair[b]
Li$_{.62}$Pb$_{.38}$ (eutectic)	464	464-464	Low	0.32	Fair[b]
Oxides					
Li$_2$O	~ 1700	410-910	Very High	0.93	Good
α-LiAlO$_2$[a]	1610	500-850	Moderate	0.27	Fair[b]
Li$_2$ZrO$_3$	1615	[c] 420-860	High?	~ 0.33	Fair[b]
Li$_2$SiO$_3$	1200	[c] 420-610	Moderate	0.36	Fair[b]
Li$_4$SiO$_4$	1250	[c] 420-640	Moderate	0.54	Fair[b]
Li$_2$TiO$_3$	1550	[c] 420-820	Moderate	0.33	Poor[b]

[a] α-γ phase transition at ~ 930°C.

[b] Neutron multiplier required.

[c] Estimated.

microstructure be preserved. There is serious concern about several possible effects:

1. sintering and pore closure (could prevent T_2O gas release),

2. tritium trapping,

3. swelling,

4. restructuring/grain growth, and

5. burnup of Li.

Radiation effects upon solid breeders are not well understood at this time, but such effects could impact upon the viability of solid breeders.

Summary

Anticipated performance of tritium breeding materials was discussed above. Given the constraint that the breeder material must not react vigorously with the coolant, a relatively small number of breeder/coolant combinations (Table 4) appear to be potentially viable. The major concerns associated with liquid metals are MHD effects, corrosion/mass transfer, and chemical reactivity. However, tritium recovery and radiation effects are not believed to be serious problems for the liquids. By contrast, the major concerns associated with solids are tritium recovery and radiation effects. Reactivity and corrosion are of much less concern for solids, particularly the ternary oxides. At the present time, not one of the combinations listed in Table 4 has been developed to a point where one could predict with any degree of confidence that it would perform satisfactorily in a fusion reactor.

Table 4. Plausible Breeder/Coolant Combinations

Breeding Materials	Combinations		Key Issues
	Breeder	Coolant	
I. Liquid Breeders			
A. Li	Li	Li	MHD Effects
	Li	He	Chemical Reactivity with Air or Water
B. $Li_{.17}Pb_{.83}$ ($Li_{.1}Pb_{.4}Pi_{.5}$)	$Li_{.17}Pb_{.83}$	$Li_{.17}Pb_{.83}$	Peak Operating Temperature
	$Li_{.17}Pb_{.83}$	He, H_2O	Corrosion
			Mass Transfer
II. Solid Alloys and Intermetallics			
A. Solid Alloys (Li_7Pb_2, Li_3Bi, LiAl)	Li_7Pb_2	He	Temperature Range
			Tritium Recovery
			Blanket Engineering
B. $Li_{.62}Pb_{.38}$ (Eutectic, m.p. 737 K)	$Li_{.62}Pb_{.38}$	He	Chemical Reactivity with Water
			Radiation Effects
III. Oxides			
A. Li_2O	Li_2O	He, H_2O	Chemical Reactivity with Water
	Li_2O	Li_2O	Tritium Recovery
B. Ternary Oxides: (α-$LiAlO_2$, γ-$LiAlO_2$, Li_2SiO_3, Li_4SiO_4, Li_2TiO_3, Li_2ZrO_3)	$Li_xM_yO_z$	He, H_2O	Radiation Effects
			Blanket Engineering
			Phase Stability
			Neutron Multiplier (Group B)
			Corrosion

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