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W. R. Grimes
Stanley Cantor

OAK RIDGE NATIONAL LABORATORY
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REACTOR CHEMISTRY DIVISION

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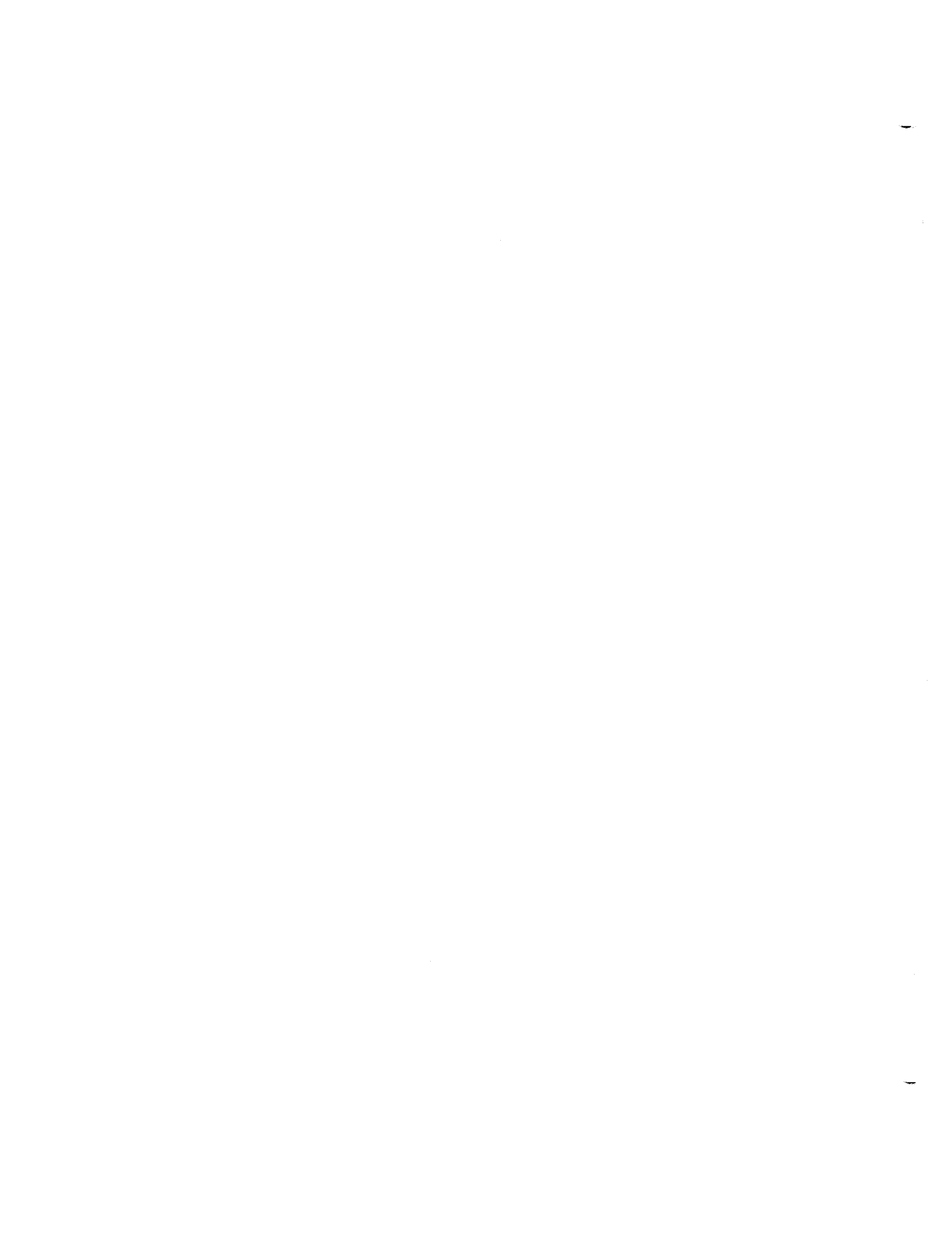
DECEMBER 1972

OAK RIDGE NATIONAL LABORATORY
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MASTER

CONTENTS

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W. R. Grimes and Stanley Cantor

ABSTRACT

The blanket of a fusion reactor serves to absorb and transfer the energy of the fusion reaction products, and to produce the tritium necessary to refuel the reactor. This report outlines how these two functions are performed by lithium-bearing molten salts.

The strong magnetic fields may have a considerable effect on the chemical stability and a less significant effect on the fluid dynamics of a flowing salt. A salt melt flowing across a strong magnetic field induces an electric field, which in turn produces an emf between the walls of the conduit and the adjacent salt. The emf can be lessened to minor proportions by careful design--causing the salt to flow parallel to the magnetic field wherever possible and using a system of small bore tubes where the flow must cross the magnetic field. Although flow in the magnetic field parallel to the lines of force suppresses turbulence (necessary in a salt for adequate heat transfer), this effect on molten salts will be negligible owing to their low electrical conductivities.

Breeding of tritium in a molten-salt blanket is at best marginal when the lithium is in its natural isotopic abundance; however, the tritium-breeding ratio can be improved by including blanket regions of lithium or of beryllium, or by enriching the salt in lithium-6. LiF and its mixtures with BeF_2 are the best molten-salt coolants in which to breed tritium in quantities adequate for fueling a reactor. Molten $LiF-BeF_2$ is advantageous in recovering tritium since, in contact with metallic Ni, Mo, or W, virtually all of tritium is present as TF.

While fluorides are adequate heat-transfer agents and possess good radiation stability, neutronic transmutation of Be and F in the salt can lead to corrosion unless a redox buffer (analogous to U^{3+}/U^{4+} in a fission reactor) is included in the melt.

In a blanket which has two coolants, one being metallic lithium, salts other than $LiF-BeF_2$ could be considered. For example, LiCl-KCl, melting at $354°C$, may be adequate as vacuum wall coolant and as the fluid to transport heat to the steam system of the reactor.

INTRODUCTION

A controlled thermonuclear reactor (CTR) that fuses deuterons with tritons yields 17.6 Mev per fusion event mostly in the form of very energetic neutrons. Such a device requires a blanket system, a more or less complex composite of several materials, capable of performing at least two functions. These functions are (a) absorption of the energy carried by the energetic fusion reaction products and transfer of heat generated in the blanket to the power-producing portion of the reactor, and (b) generation of tritium, in a manner such as to enable its ready recovery, to replace that consumed in the fusion reaction. The first of these functions certainly requires a suitable heat transfer fluid. The second requires that the blanket contain a sufficiency of lithium, from which tritium can be effectively bred. Though these two functions are separable in principle,[1] there is probably a considerable advantage, other than elegance, if the coolant fluid can be a liquid with sufficient lithium to sustain the required tritium production.

The coolant fluid must meet several criteria. These are generally similar to the requirements imposed on molten salts as fuels for fission reactor fuels (1). It must not adversely interact with neutrons necessary for breeding of tritium. It must be a good heat transfer fluid and its heat transfer and hydrodynamic behavior must (for most applications) be adequate in the presence of large magnetic fields. It must be non-corrosive toward metals of construction in the blanket region, the pumps, and the power generation equipment. It must be suitably stable to the intense radiation fields within the blanket and it must not react violently if, upon failure of the heat transfer equipment, it is inadvertently mixed with the power-generating fluid (steam, potassium, etc.). It should, in addition, possess a relatively low vapor pressure to minimize stresses on the blanket structure, and it should be compatible with auxiliary blanket subsystems such as neutron moderators (graphites) or neutron multipliers. Finally, this fluid should also be capable of efficient

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[1]It is possible, at present, to visualize a blanket in which tritium is bred in (essentially) stationary bodies of lithium metal, lithium alloys, or lithium salts, with the necessary cooling accomplished by another fluid--molten sodium, pressurized helium, gas, or molten salt.

generation of tritium and it should be of a nature such as to permit easy recovery of the tritium for return to the fusion cycle.

This paper attempts to assess the potential of molten salts as blanket fluids in controlled thermonuclear reactors. As matters stand at present, several entirely different types of CTRs are potentially feasible. This fact coupled with the possibility (even the likelihood) that the coolant and the breeding functions are separable makes it impossible to examine in a single document all the credible combinations.

We have chosen instead, in what we hope will prove to be a continuing examination of the overall problem, to do the following:

1. To assess the problems for the most difficult case, a single molten salt coolant and breeder blanket in a closed magnetic field device. We will use molten Li_2BeF_4, probably the most studied and best understood of the possible candidates, for this assessment in a hypothetical Tokamak; a Stellerator would be equivalent in nearly every regard.

2. To compare several molten salts for this and other less demanding possible CTR uses,

3. To consider, briefly, application of molten salts for a laser-powered fusion device whose problems are quite different, and finally

4. To compare and contrast these molten salts with molten lithium as the blanket fluid.

BEHAVIOR OF Li_2BeF_4 IN A HYPOTHETICAL CTR

The most difficult problems which must be surmounted by a molten salt in a CTR blanket system are, almost certainly, those imposed if the salt must serve as the single-fluid blanket-coolant for a closed magnetic field (and essentially steady state) CTR. The precise magnitude of these problems will depend upon the detailed design of the device--its power level, size, temperature and temperature differential, and coolant passage patterns within the blanket region. To illustrate the nature and general magnitude of the problems we have chosen a toroidal CTR, blanketed and cooled with molten Li_2BeF_4, of the size and characteristics shown in Table 1.[2] The blanket dimensions are essentially those proposed by

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[2]This design is a hybrid from sources indicated in the text. It is presented only for illustration of several problems to be described.

Fraas (2) for a lithium-metal cooled device; the magnetic fields are those indicated in an Oak Ridge National Laboratory study (3); flow rate and temperature differential are scaled from a proposed 1000 MW(e) molten salt fission reactor (4).

It is clear from Table 1, that the efficiency of the device (ca 45%) presupposes temperatures above 700°C for the coolant outlet and that mean residence time of the fluid within the active blanket region is (since only a small fraction of the inventory is in the pumps, piping, and heat exchangers) approximately five minutes. It is assumed for this particular device that the Li_2BeF_4 is pumped through the blanket region (and through the enormous magnetic field) at 120 cubic feet per second and directly to equipment for generation of high quality steam. These assumptions, as to pumping rates, ΔT, and steam generation compatibility, are not unlike those proposed for molten salt breeder (fission) reactors (MSBRs) (4).

Table 1. Characteristics of a Hypothetical
Controlled Thermonuclear Reactor

Power	2250 Mw(th) [1000 MW(e)]
Major Diameter	21 meters
Minor Diameters:	
First Wall	7 meters
Blanket Region	9 meters
Shield Region	11 meters
Magnetic Fields:	
Maximum Toroidal Field at the Coils	80 kgauss
Toroidal Field at the Center of the Plasma	37 kgauss
Poloidal Field (pulsed) in the Plasma	7 kgauss
Blanket Characteristics:	
Salt (Li_2BeF_4)	60%
Graphite (or Be)	40%
Inventory of Li_2BeF_4	2×10^6 kg
Flow Rate	4×10^5 kg/min (120 ft^3/sec)
ΔT	167°C (300°F)

It is worthy of note that the <u>mean</u> radiation load on the Li_2BeF_4
(ca 1.1 watts/gram) is more than 10-fold below that proposed for MSBRs.
This comparison is, however, deceptively favorable toward MSBRs in that
these reactors can assure moderately uniform radiation levels within
their fuels while CTRs cannot do so for their blankets. It is likely
that radiation densities in CTR blanket near the plasma-confinement
(first) wall will be larger than those proposed for the MSBR. This CTR
radiation density is not, however, likely to approach the maximum radia-
tion density at which molten salts have been tested (5).

Such a device as that described above will, on the other hand, pose
several problems for the Li_2BeF_4. Some of these problems are similar
and some are quite different from those posed by use as fuel solvents in
fission reactors. These problems are defined and discussed in the
following sections.

Effects of Strong Magnetic Fields

Pumping a conducting fluid into (across the magnetic field lines of)
a closed magnetic field device poses a problem. For a liquid metal this
problem manifests itself as a large pumping power loss due to magneti-
cally induced turbulence; for molten salts the effect appears as chemical
destabilization. After the fluid is within the magnetic field one can,
in principle (and perhaps, with considerable difficulty, in practice)
make the flow channels conform closely to the magnetic lines of force.
In that case the magnetic field may exert a pronounced effect upon the
fluid dynamics of the flowing stream.

<u>Effects on Chemical Stability</u>

From electromagnetic theory it is known that the electric field
induced in a conducting fluid crossing a magnetic field is given by
the cross-product of fluid velocity and magnetic field:

$$\vec{E} = \vec{V} \times \vec{B}$$

Molten Li_2BeF_4 (or any conducting fluid) flowing at 10 meters/
second in a pipe of 5 cm diameter with its axis aligned perpendicular to
the lines of force of an 80 kgauss (8 volt-sec/meter^2) magnetic field
will have induced, at right angles to both the magnetic field and the

flow direction, a potential difference of 4 volts between the salt and the pipe wall. Potential differences of such magnitudes are clearly intolerable; though LiF and BeF_2 are both very stable compounds (6) an induced voltage such as this (equivalent to destabilization by 92 kcal/mole) would make these compounds quite corrosive to the metallic tube walls.

Homeyer (7), who seems to have been the first to consider such electrolytic corrosion in a CTR blanket system, noted that such corrosion should be alleviated by (a) reducing fluid velocities perpendicular to the magnetic field, and (b) using a series of parallel pipes to reduce the pipe dimension where flow across the magnetic field lines are necessary.

If, for example, the 120 ft^3/sec flow of Li_2BeF_4 required of our hypothetical 1000 MW(e) CTR were supplied at a flow rate of 4 meters/sec through pipes of 4 cm diameter perpendicular to the 25 kgauss field[3] the emf induced in each pipe would be 0.4 volts; some 675 pipes would be required. If these pipes penetrated the field at 30° to the field lines this emf would be reduced to 0.2 volts. These conditions would seem to be tolerable. Alternatively, by supplying external cooling to each pipe, as it crosses the magnetic field lines, so as to form a poorly conducting layer of frozen salt on the inner pipe wall, it may be possible to reduce the number of pipes and to somewhat increase their size. Periodic replacement of corroded pipe sections might also be considered, since they are located at the periphery of the torus.[4]

Plasma stability in a reactor such as this requires a pulsed poloidal magnetic field transverse to the main field and, accordingly, perpendicular to the fluid flow parallel to the fuel lines of the main (toroidal) field. Chemical effects of this poloidal field (which may reach 7 kgauss) may not be trivial, but they would seem, in general, to be tolerable.

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[3]This is roughly the maximum field between the coils (3) in a pipe entering the outer edge of the torus.

[4]Other alternatives exist. It should be possible to penetrate the field by shafts of mechanical pumps to permit use of Li_2BeF_4 to transfer heat to (for example) a boiling potassium cycle within the magnetic field. Such alternatives are beyond the scope of this document.

It is clear that the problems outlined above deserve experimental study especially in the area of kinetics of de- and re-stabilization of real fluids upon passage through intense magnetic fields.

Effects on Fluid Dynamics

To avoid induced electric fields as discussed above, flow of the blanket fluid within the torus will be aligned with the magnetic lines of force. However, in that case the magnetic field will exert a force opposed to eddies within the fluid and will tend to damp turbulent flow. Heat loadings in the blanket structure of a CTR, and especially at the first (plasma-confining) wall, will be very large, and molten Li_2BeF_4 must develop turbulent flow if it is to cool this wall effectively. It is important, therefore, to assess this damping effect of the field on turbulent flow in the salt.

No experimental study of magnetic damping in molten salt has been reported, but experiments with liquid mercury have been performed (8,9). These experiments with mercury appear to provide a means of estimating the Reynolds number at which the transition from laminar to turbulent flow occurs. A dimensionless quantity which characterizes the magnetic forces that affect flow is called the Hartmann number (M), and is defined:

$$M \equiv B\ell(\sigma/\eta)^{\frac{1}{2}}$$

where   B is the applied field,
       ℓ is a characteristic length, usually the half-width of the flow channel,
       σ is the fluid's electrical conductivity, and
       η is the viscosity

In the absence of a magnetic field the laminar-turbulent transition occurs at Reynolds number (R_o) of about 2200; in the presence of the field this transition occurs at a higher Reynolds number (R_t). This increase (more commonly the ratio (R_t/R_o) is a function of the Hartman number (M). Three such correlation functions have been published (8,9,10).

For Li_2BeF_4 at $600^{\circ}C$, the electrical conductivity (11) is 220 (ohm-meter)$^{-1}$, the viscosity is 8×10^{-3} kg/(sec-meter). Accordingly, for this material in a 80 kgauss (8 webers/meter^2) field and assuming a 6 cm (3 cm half-width) channel the value of the Hartman number is 40. If we apply each of the three correlations (8,9,10) developed from experiments with mercury to a fluid of

$$M = 40$$

we find the transition Reynolds number should rise from $R_o = 2200$ to $R_t = 2720$, 3740, and 2400, respectively. The effect of magnetic field on this fluid is, therefore, predicted to be relatively small.

These three values for R_t are very much less than values estimated for flows in the first wall region. As an approximation let us assume that to cool the vacuum wall we require 40 ft^3 per second of salt (one third the quantity required to cool the entire blanket) and that this salt flows through a 6 cm wide annulus around the 7 meter diameter first wall. The linear velocity of salt is 0.85 meters/sec, the density (11) (at 600°C) of Li_2BeF_4 is 1990 kg/meter^3 and the viscosity is 8 centipoise. The Reynolds number under these conditions is about 13,000.

These data strongly suggest that molten Li_2BeF_4 can be made to flow turbulently within the blanket system, but direct experiments with molten salts would certainly appear desirable.[5]

Any transverse magnetic field will also act to suppress turbulence. Hoffman and Carlson (10) propose the formula, $R_t = 500M$ for calculating the transition Reynolds number of Mercury flowing transverse to the magnetic field. The same formula applied to Li_2BeF_4 at 600°C flowing in a 6-cm thick channel transverse to 8 kgauss (the poloidal field strength) yields

$$R_t = 500M = 2000.$$

This result suggests that an 8 kgauss poloidal field in a toroidal reactor may not affect turbulence in Li_2BeF_4; at most, the suppression of turbulence by this field will be comparable to the effects of the toroidal field.

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[5]Metallic lithium is expected to be quite different. The Hartman number for Li is 12,000 for the condition where that of Li_2BeF_4 is 40; the corresponding R_t for lithium is, accordingly, above 700,000. It would appear that Li will be constrained to laminar flow; it seems likely, however, that (because of its good thermal conductivity) it can adequately cool the first wall.

Production of Tritium

It is obviously necessary to use the neutrons produced in fusion to breed the tritium required to fuel a D-T reactor. If the only sources of tritium were today's transmutation facilities then the fuel cost alone would be about 1.2 cents per kilowatt-hour.[6]

The only practical neutron reactions which will yield tritium sufficient for the needs of a D-T fusion reactor are $^6Li(n,\alpha)T$ and $^7Li(n,\alpha n')T$. The latter is a high-energy reaction with a threshold at 2.5 MeV. Neutron-capture cross sections of ^6Li become significant only at energies of 0.5 MeV and less. The ^7Li reaction is particularly favorable since the product neutron (n') can react with ^6Li to yield a second tritium atom, but because the blanket contains elements which scatter and absorb high-energy neutrons, the production of tritium from ^7Li is not very efficient. When the lithium in the blanket is in natural isotopic abundance (92.58% ^7Li, 7.42% ^6Li), the greater fraction of tritium is produced from ^6Li.

Several studies have been reported concerning the breeding of tritium in fusion reactor blankets (12-16). In cases where the lithium is in natural isotopic abundance, tritium-breeding ratios clearly greater than unity are calculated for lithium metal blankets; the breeding ratios for lithium salts are distinctly less favorable. Table 2 presents some very recent calculations for four salts and for lithium metal all in the same blanket configuration. Some of the neutron cross sections, particularly those for the n,α, and n,n'γ reactions of fluorine (17), are uncertain. The values in Table 2 are, therefore, more useful for comparison than for accurate prediction of tritium production;[7] uncertainties of perhaps 10% in the calculated breeding ratios are possible.

Three options, either alone or in combination, can be considered for upgrading tritium production when the breeding ratio is marginal as appears to be the case for Li_2BeF_4.[8] These are:

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[6]Based on a tritium cost of 10 cents/curie and 22 MeV/fusion utilized in a plant operating at 40% thermal efficiency.

[7]It should be noted that Blow, et al. (14) calculate a tritium breeding ratio of 1.027 for Li_2BeF_4 in a blanket assembly similar to that of Table 2.

[8]Possible use of LiF, LiCl, and Li_2CO_3 is discussed briefly in a subsequent section of this report.

Table 2. Tritium Breeding Calculations[a,b]

Coolant[d]		Breeding Ratio[c] from		Total Breeding Ratio
		^6Li	^7Li	
LiF	$(850^\circ C)^e$	0.80_4	0.24_4	1.05
Li_2BeF_4	$(850^\circ C)$	0.78_5	0.14_4	0.93
Li_2CO_3	$(900^\circ K)$	0.64_2	0.16_7	0.81
LiCl	$(900^\circ K)$	0.61_4	0.13_2	0.75
Li	$(850^\circ C$	0.98_2	0.45_4	1.44

[a]D. Steiner, Oak Ridge National Laboratory, personal communication, March 1972.

[b]Blanket Configuration: (1) first wall - 0.5 cm Nb, (2) 94% coolant, 6% Nb - 3 cm, (3) second wall - 0.5 cm Nb, (4) 94% coolant, 6% Nb - 60 cm, (5) graphite - 30 cm, (6) 94% coolant, 6% Nb - 6 cm.

[c]Defined as tritium atom produced per fusion neutron incident on the first wall.

[d]Lithium in natural isotopic abundance.

[e]Temperature at which atom densities were calculated.

(a) design of blanket to include a region of metallic lithium,

(b) increasing Be content of the blanket by adding a region of Be or Be_2C to increase neutron multiplication and to provide more ^6Li

$$^9_4Be(n,\alpha)\ ^6_2He\ \xrightarrow{\ 0.8\ sec\ }\ ^6_3Li,\ and$$
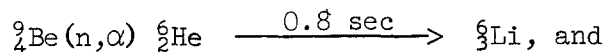
(c) modest enrichment of the blanket material in ^6Li.

The second option was briefly treated by Bell (16) who showed that if a blanket region (40 cm thick adjoining a 1 cm first wall of molybdenum) were changed from Li_2BeF_4 to an equal thickness of Be and Li_2BeF_4, the tritium-breeding ratio would increase from 0.95 to 1.50.

The third option has also received some attention. Impink (13) reported that small increases in ^6Li enrichment of the Li_2BeF_4 blanket led to modest gains in breeding ratio. For example, increasing the ^6Li isotopic fraction to 0.2 in a 6.25-cm thick coolant region next to the first wall improves the total breeding ratio by about 3%. Although

enrichment costs are high, these costs would be partly offset by improved shielding of the magnet coils and by reduced radiation damage to the first wall through reduced resonance capture (13).

In light of present knowledge of the pertinent cross sections, it appears that the breeding capability of Li_2BeF_4 is marginal in devices such as our hypothetical torus. This material would, therefore, probably need to be augmented by one of the methods indicated above, or by other means. It is clear that better cross section data are needed so that this point can be decided.
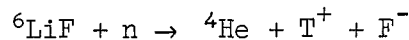
Recovery of Tritium

Approximately 270 grams of tritium are consumed per day by fusion in a 2250 MW(t) D-T reactor. Slightly more than this, or approximately 300 grams per day, must be produced and recovered; this corresponds to some 50 moles of T_2 or to 100 moles of TF per day. In our hypothetical CTR the mean residence time of the fluid in the blanket is 5 minutes per cycle. Some 0.174 moles of T_2, or 0.348 moles TF is, accordingly, produced in the Li_2BeF_4 in this interval. If the fluid entering the blanket contained no tritium species the fluid emerging from the blanket will contain about 1.74×10^{-7} moles T_2 (or alternatively about 3.48×10^{-7} moles TF) per liter if complete homogeneity is assumed. The problems in recovery and management of the tritium depends significantly on whether the material exists as T_2 or as TF. These two situations, and the extent to which the mode of tritium behavior can be controlled, are briefly described in the following.

Solubility of H_2 in molten Li_2BeF_4 has been shown to increase linearly with pressure of H_2; at $1000°K$, the solubility should be near 7×10^{-5} moles H_2 per liter of salt per atmosphere of H_2 (18). No studies of tritium solubility have been reported. If the bred tritium occurs as T_2, and if the solubility behavior of T_2 and H_2 are similar, the emerging blanket fluid carries T_2 (generated during its pass through the blanket) equivalent to a saturating pressure of about 2.5×10^{-3} atmospheres. Equilibration of the emerging salt with a relatively small volume of inert gas will result in stripping of a very large fraction of this dissolved T_2 from the salt. It is clear, however, that this process

(with 120 ft^3 of salt and, for example, 1 ft^3 of He) will be difficult to engineer, and, moreover, that diffusivity of T$_2$ at such effective partial pressures through hot metal surfaces will pose problems.
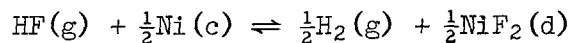
The solubility of HF in molten Li$_2$BeF$_4$ also depends linearly on pressure of the solute gas, and its Henry's law constant is 10^{-2} moles HF per liter of salt per atmosphere HF at 1000°K (19). The TF produced during each cycle of coolant through the blanket region will correspond to about 3.5 x 10^{-7} moles TF per liter of Li$_2$BeF$_4$; this is equivalent to a saturation pressure of about 3.5 x 10^{-5} atmospheres of TF. The TF will be more difficult to strip from the salt than will T$_2$, but TF will not diffuse through the metal walls. If its reaction with the metal walls can be sufficiently minimized, the TF concentration can be allowed to increase and the rate of processing the blanket fluid can be correspondingly reduced. If, for example, the TF can be allowed to concentrate until its pressure is 10^{-3} atmospheres, sparging of perhaps 5 ft^3/sec of the fluid with helium should suffice for effective recovery of the bred tritium. It is, accordingly, worthwhile to examine whether the bred tritium can reasonably be maintained as TF.

Tritium produced, for example, from
$$^6\text{LiF} + n \rightarrow \ ^4\text{He} + \text{T}^+ + \text{F}^-$$
is, in principle, born as an oxidized species. The tolerable concentration of TF, or of any other oxidized species, will, of course, be limited by the extent to which corrosive reaction with the CTR metal can occur. If the containment metal is sufficiently inert, useful concentrations of TF can be maintained without appreciable reaction.

By way of illustration, let us examine the reaction of HF with nickel,
$$\text{HF(g)} + \tfrac{1}{2}\text{Ni(c)} \rightleftharpoons \tfrac{1}{2}\text{H}_2\text{(g)} + \tfrac{1}{2}\text{NiF}_2\text{(d)}$$
where (g), (c), and (d) indicate, respectively, that the species is gaseous, crystalline solid, or dissolved in molten Li$_2$BeF$_4$. From the data of Table 3 ΔG° = 10.9 kcal for this reaction at 1000°K. The equilibrium constant is given by
$$K = \frac{N_{\text{NiF}_2}^{\frac{1}{2}} \ P_{\text{H}_2}^{\frac{1}{2}}}{a_{\text{Ni}}^{\frac{1}{2}} \ P_{\text{HF}}} = 4 \times 10^{-3}$$

where N is the mole fraction of dissolved NiF_2, a is the activity of nickel (unity in this case), and P is the partial pressure of the designated gaseous species. If we set $N_{NiF_2} = 3.2 \times 10^{-6}$ (equivalent to 6 parts per million of Ni^{2+}, a value that seems likely to be tolerable) we calculate pressures of H_2 of 6×10^{-9} atmospheres and 5×10^{-6} atmospheres, respectively, in equilibrium with HF pressures of 3.5×10^{-5} and 10^{-3} atmospheres. These results suggest that if the CTR metal were Ni a very large fraction of the tritium could be maintained as TF and stripped as such.

Examination of Table 3 suggests that the situation may be even more favorable for molybdenum and, perhaps, for tungsten as the containment metal. However, if the containment metal were iron, chromium, niobium, tantalum, titanium, or, probably, vanadium the tritium must, of necessity, be stripped and handled as T_2. If one of these more reactive metals proves necessary as the CTR material, some way of preventing corrosion due to

$$xTF + M \rightarrow MF_x + \frac{x}{2}T_2$$
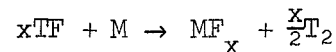
must be provided. This would seem to be possible by incorporation of a redox buffer (described in more detail in the subsequent section) in the molten Li_2BeF_4.

At this stage in the technology of fusion reactors one should probably not dismiss the possibility of using stainless steel or a chromium-containing nickel-based alloy at temperatures at or below $1000^{\circ}K$. Such materials can, perhaps, be coated with molybdenum, tungsten, or nickel, by electrodeposition (22) or by plasma spraying (23).

Chemical Transmutations

Several types of chemical transmutations will occur in molten Li_2BeF_4 in its service as the blanket fluid in a fusion reactor. The most important of these, and means for maintenance of the blanket to minimize or avoid their deleterious effects, are the following:

Transmutation of lithium is, of course, essential to production of tritium. The overall reactions can be represented as

$$^7LiF + n \rightarrow \frac{4}{2}He + TF + n', \text{ and}$$
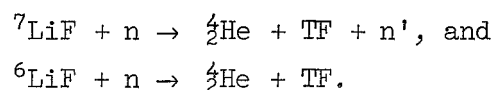$$^6LiF + n \rightarrow \frac{4}{2}He + TF.$$
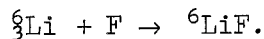
Table 3. Free Energies of Formation of Fluorides

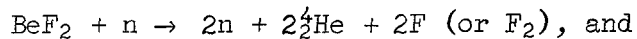|	$\Delta G^f_{1000°K}$ (kcal/g-atom of fluorine)	Reference
MoF_6 (g)	-50.2	(20)
WF_6 (g)	-56.8	(6)
NiF_2 (d)[a]	~55.3[a]	(20)
VF_5 (g)	-58[b]	(21)
VF_4 (cr)	-66[b]	(21)
HF (g)	-66.2	(6)
FeF_2 (d)[a]	-66.5[a]	(20)
NbF_5 (g)	-72.5	(20)
CrF_2 (d)[a]	-75.2[a]	(20)
TaF_5 (g)	-82.2	(20)
TiF_4 (g)	-85.4	(6)
LiF (ℓ)	-125.2[a]	(20)
BeF_2 (ℓ)	-106.9[a]	(20)

[a]Standard free energy of formation in molten Li_2BeF_4.

[b]Estimated from the relation, $\Delta G^f_{1000} = \Delta H^f_{298} - 1000\ (\Sigma\ S^o_{298})$ taking ΔH^f_{298} and most S^o_{298} from NBS Technical Notes 270-3,4,5 (21). Other S^o_{298} estimated from analogous compounds.

These reactions are not inherently oxidizing or reducing, though, as described in the previous section, the generated TF can oxidize reactive structural metals to form metal fluorides which will dissolve in the melt.

Transmutation of beryllium (as BeF_2 in Li_2BeF_4) leads to corrosion of any system metal since disappearance of Be^{2+} is equivalent to release of fluorine. The two reactions may be represented as:

$$BeF_2 + n \rightarrow\ 2n + 2^4_2He + 2F\ (or\ F_2),\ and$$

$$BeF_2 + n \rightarrow\ ^4_2He + ^6_2He + 2F,\ followed\ by$$

$$^6_2He \xrightarrow{\ 0.8\ sec\ half\ life\ }\ ^6_3Li,\ and$$

$$^6_3Li + F \rightarrow\ ^6LiF.$$
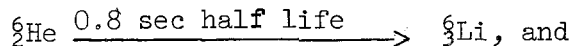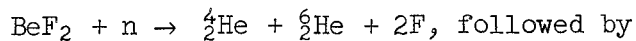
In our 2250 MW reactor, these reactions yield, respectively, the equivalent of 500 g and 70 g of fluorine per day. This problem is generally similar to that encountered in fission of uranium (as UF_4) in the MSRE (1);

it is clearly necessary to provide a redox buffer in the molten salt
(the UF_3-UF_4 couple does this in fission reactors), capable of oxidizing
F^O to F^-. It is also necessary, if Ni, Mo, or W constitutes the container
system, that this redox buffer be consistent with maintenance of the
tritium as TF. The couple

$$Ce^{3+} \rightleftharpoons Ce^{4+}$$

may possibly serve this function. If, for example, the concentration
of cerium in the melt is set at 10^{-4} mole fraction the blanket will con-
tain 6×10^3 mole of Ce^{3+} + Ce^{4+}, and the Ce^{3+}/Ce^{4+} ratio would require
chemical adjustment on a cycle time of many days. If, on the other hand,
the container metal is Nb (or some other metal which will reduce TF in
dilution solution) the redox couple must be chosen so as to be consider-
ably more reducing. It must deal with the F_2 generated by transmutation
of beryllium but it must also reduce the 100 moles per day of TF produced
by transmutation in the LiF. Such a buffer system would require adjust-
ment on a cycle of a few days.

In addition, transmutation of fluorine occurs upon capture of
neutrons of energy above about 3 MeV. This reaction may be represented
by

$$^{19}_{9}F^- + n \rightarrow ^{16}_{7}N^- + ^{4}_{2}He.$$
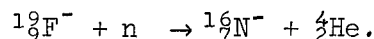
This nitrogen isotope decays, with a 7.3 sec. half-life, to an oxygen
isotope

$$^{16}N^- \rightarrow ^{16}O + \beta^-,$$
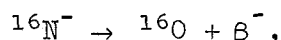
and the result is probably, although the mechanism may be complex, grow-
in of O^{2-}. The absolute quantity of ^{16}N formed by this reaction is
relatively uncertain; it is estimated to be, within a factor of three,
120 grams/day. The very short half-life of this isotope guarantees that
all the ^{16}N decays within the CTR blanket. The concentration of ^{16}N,
in whatever chemical form, within the Li_2BeF_4 cannot exceed 1.1 parts
in 10^{11}. However, some fraction of this material will react with the
CTR containment metal; decay of this isotope will lead to formation of
metal oxide in the CTR metal. This may, especially if it concentrates
within the grain boundaries, prove troublesome. If all the $^{16}N^-$ decayed
within the blanket salt, the oxide concentration of our hypothetical CTR
would increase about 60 parts per billion per day. Since 10 to 50

parts per million of oxide is almost certainly tolerable, a process for removal of oxide on a cycle time of several months to several years should suffice.

Finally, it should be noted that the transmutation reactions shown all generate He. For the hypothetical CTR the daily production of helium is about 125 gram atoms or nearly 100 standard cubic feet. Helium is relatively insoluble in molten Li_2BeF_4 (24); the solubility at $1000^{\circ}K$ is 1.7×10^{-4} moles He per liter salt per atmosphere. Helium produced per pass of blanket salt corresponds to a saturation pressure of 2.6×10^{-3} atmospheres. If no sparging were attempted the helium pressure would reach 1 atmosphere in about 30 hours.

Compatibility of Li_2BeF_4 with CTR Metals and Moderators

As indicated in Table 3 above, LiF and BeF_2 in molten Li_2BeF_4 are very stable materials. Both are much more stable than the structural metal fluorides; consequently, corrosion due to chemical reactions with these major blanket constituents should prove minimal. Indeed, experience with the Molten Salt Reactor Experiment (25) has shown negligible corrosion by this fluid on a nickel-base alloy (Hastelloy N). However, such salts are excellent fluxes for metallic oxides and halides, and films of such substances afford no protection against oxidizing agents carried by such melts; accordingly, as described above, HF (or TF) may react with the containment metal, and impurity ions such as Ni^{2+} will react with metallic iron or chromium in the container metal (1).

Melts such as Li_2BeF_4 are chemically inert toward, and do not wet, graphite (1). However, the possibility that such salts will transfer graphite and carburize metals such as Mo or Nb cannot be discounted. It is not likely that a system built of Mo, Nb, or V can use molten Li_2BeF_4 and unclad graphite without adverse interactions. Similarly, metallic Be cannot react appreciably with Li_2BeF_4 (but the Be could certainly react with TF or with the Ce^{3+}/Ce^{4+} couple proposed as a redox buffer in the system). Any real use of metallic Be as a neutron multiplier in the blanket system, therefore, presupposes that the Be is clad with an inert metal.

Other processes which could conceivably give rise to corrosion can be dismissed as highly improbable. Direct dissolution of structural metals in Li_2BeF_4 has never been observed. Salt decomposition caused by the slowing down of energetic particles should not lead to corrosion provided that the salt is kept at elevated temperatures. Experience gained in the Molten Salt Reactor Experiment (26) and in an extensive in-pile radiation testing program showed that as long as the temperature was greater than $150^{\circ}C$ (27), radiolytic decomposition was of no importance to corrosion of structural metals or graphite.

Compatibility with Steam, Air, and Liquid Metals

In any system of heat-exchangers and hot flowing liquids, there exists a real and finite probability that leaks will occur. In this section we examine the consequences of leaks and intermixing of other fluids and Li_2BeF_4.

The reaction of steam with Li_2BeF_4 yields HF and BeO

$$H_2O(g) + BeF_2(\ell) \rightleftharpoons BeO(c) + 2HF(g)$$
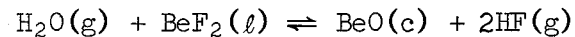
though the reaction is not particularly exothermic. Both H_2O and HF are likely to corrode the metal in contact with the salt; corrosion-product fluorides will dissolve or be otherwise carried by the salt. Since BeO is only very slightly soluble (125 ppm at $500^{\circ}C$) in Li_2BeF_4 (28), a large in-leakage of steam would soon lead to the precipitation of BeO in the salt circuit. Leakage of air into LiF or Li_2BeF_4 will have troublesome, but not hazardous, consequences. Dry air will not react directly with either salt; however, air oxidation of surfaces in contact with the salt will result in dissolution by the salt and, if continued, in ultimate precipitation of BeO. Moisture in the air will also react, as does steam, with Li_2BeF_4. The molten Li_2BeF_4 can, if necessary, be freed of oxide by treatment at elevated temperatures with anhydrous HF (29).

In some CTR designs suggested in a subsequent section of this paper, Li_2BeF_4 (or other salt) could inadvertently be mixed with liquid alkali metals. From Li_2BeF_4, metallic Li, Na, or K react to precipitate Be metal, but the reaction is not highly exothermic.

In general, although inadvertent mixing of Li_2BeF_4 (or most other molten salts) with other CTR fluids would prove troublesome, such mixing would not lead to violent or explosive reactions.

CHOICE OF MOST PROMISING SALTS

In this section we attempt to answer two questions. These are: (a) if tritium must be bred in the blanket-coolant which lithium-bearing salt is best, and (b) if the coolant and breeding function of the blanket can be separated which are the most promising molten salt coolants?

In answer to the first question, it must be conceded that obtaining breeding ratios greater than unity with molten salts alone may pose a real difficulty. Table 2 above suggests that LiCl and Li_2CO_3 show, in reasonable (though not optimized) blanket configurations, breeding ratios that are unsatisfactory. Breeding ratios have also been calculated for $LiNO_2$ (13) and $LiNO_3$ (15); the results tend to be quite unfavorable. Impink (13), for example, obtained the value 0.82 for $LiNO_2$.[9] No calculations appear to have been made for Li_2SO_4, but the high cross sections for $S(n,\alpha)$ and $S(n,p)$ reactions almost certainly will reduce the breeding ratio below that for Li_2CO_3. Moreover, $LiNO_2$ and $LiNO_3$ lack the thermal stability required of truly high temperature coolants, and Li_2CO_3 and Li_2SO_4 will oxidize many CTR structural materials.

Lithium hydroxide seems to be eliminated, even if (as is unlikely) its properties are otherwise satisfactory, because its hydrogen would excessively dilute the bred tritium. Lithium oxide (Li_2O) has a lithium density nearly 50% above that of metallic lithium, but its melting point of nearly $1470^{\circ}C$ (6) eliminates it as a major constituent of a blanket fluid. Lithium chloride melts at $610^{\circ}C$ (6) and should be reasonably compatible with CTR metals, but its breeding ratio (see Table 2) appears inferior.

The salt with the most favorable breeding ratio is LiF. This salt is inert toward graphite and to metals under consideration for the blanket structure. The major drawback of LiF is its melting point of $848^{\circ}C$.

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[9] In the same configuration he calculated 1.15 for Li_2BeF_4.

Because of this high melting temperature, LiF cannot be used to transfer
heat to the steam system of the reactor. If the blanket region were oper-
ated at very high temperatures (>900°C), then LiF could be used in conjunc-
tion with an intermediate heat-exchange medium--liquid Na, a lower melting
salt, or perhaps a boiling alkali metal system. The melting point of LiF
can be substantially lowered by many solutes; the ideal solute should lower
the melting point below 374°C[10] without affecting either the breeding gain
or the generally favorable heat-removal and chemical properties of LiF.
We know of no such solute. Dissolved Li_2O should increase the breeding
ratio slightly, but considering the probable limited solubility of Li_2O,
the melting temperature (more accurately, liquidus temperature) of LiF will
not drop below 800°C. Using AlF_3 and/or another alkali fluoride to lower
the melting temperature to ~700°C should not have dire chemical consequences,
but the breeding ratio will almost certainly suffer. The nearest approxi-
mation to an ideal solute in LiF is probably BeF_2. The phase diagram for
LiF-BeF_2 (30), presented in Fig. 1, shows that a melting temperature as low
as 363°C is available in this system. Unfortunately, the viscosity of the
melt increases with BeF_2 concentration, and mixtures with >40 mole % BeF_2
have viscosities greater than 50 centipoise at 450°C (31). The optimum
salt mixture of low melting temperature and acceptable viscosity, and with
a reasonably good tritium breeding ratio is at ~33 mole % BeF_2, correspond-
ing to the Li_2BeF_4 used for illustrative purposes in earlier sections of
this report. Decreasing the BeF_2 concentration below 33 mole % may have a
modest beneficial effect upon breeding ratio; this increase might, possibly,
offset disadvantages posed by the increased liquidus temperature and likely
changes in chemical behavior.

In summary, the answer to the first question posed above--the best
blanket coolant salt in which to breed tritium is LiF, but its melting
point of 848°C limits its usefulness only for cooling a blanket that
operates above this temperature. If the blanket coolant must also trans-
fer heat to the steam system, Li_2BeF_4, or some modest variant of this
composition, appears to be the best choice.

A partial separation of breeding and cooling functions, posed in the
second question above, has been approached by Steiner (12). He calculated

[10]The critical temperature of H_2O.

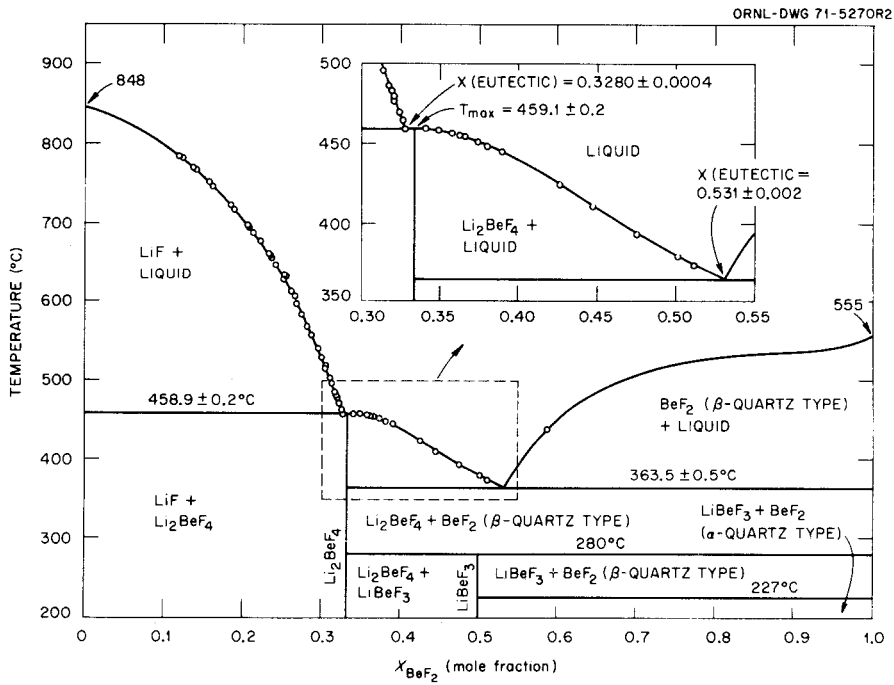Fig. 1. Phase Diagram of the System LiF-BeF$_2$.

the tritium breeding ratio for a blanket design in which Li_2BeF_4 cooled the vacuum wall and lithium metal assumed the rest of the heat-transfer function, and showed that the breeding ratio (1.22) was 8.3% less than that (1.33) for the same blanket with lithium as the sole coolant. This small loss in breeding ratio suggests that other salts, especially those melting below $374°C$, might also serve as vacuum-wall coolants. Moreover, the heat carried by the lithium might then be transferred outside of the blanket to a molten salt of the same composition as the vacuum-wall coolant. The salt streams from the vacuum wall and the lithium heat-exchanger would then be combined and pumped to the steam-raising system. The eutectic mixture of LiCl and KCl, which melts at $354°C$ (32), might be suitable for such service. The authors are presently assessing the implications of this "double-coolant" concept.

In principle, a complete separation of breeding and cooling functions might be embodied in a blanket design in which a molten salt cools the vacuum wall, a moderately thick region of quiescent lithium metal, and a graphite moderator-reflector.[11] The coolant salt for this application must have great chemical stability (to avoid deleterious destabilization in the magnetic field and to avoid corrosion of the CTR metal), sound heat transfer properties, and, preferably, a low freezing point. These specifications narrow the choice of major component to fluorides, chlorides, and oxides, of lighter alkali and alkaline earth metals which melt below $1200°C$. The list seems to contain LiF, LiCl, NaF, NaCl, Na_2O, KF, KCl, BeF_2, $MgCl_2$, and $CaCl_2$ with, perhaps, other compounds of these families as possible minor constituents of mixtures. When one adds the further requirement that the coolant not lower the breeding ratio below unity, the list of useful major components becomes smaller. The high inelastic scattering cross sections for Mg, for example, probably eliminates any substantial concentration of $MgCl_2$ from the coolant. From consideration of this list of materials it would appear that LiF is best except for its high melting point, that Li_2BeF_4 may well be the best overall choice, and

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[11]A possible rationale for such a design is that lithium, though providing a comfortable breeding ratio, cannot be made to flow turbulently within the blanket and may require excessive power in being pumped through the major magnetic field.

that, if subsequent calculations show that the breeding ratio does not suffer unduly, the ternary eutectic LiF-NaF-KF (melting point 454°C) and the binary eutectic LiCl-KCl may be suitable coolants.

Practical problems with these materials will differ in detail from those described earlier for Li_2BeF_4. Superficial examination of these problems reveal none that seem insuperable, but much experimental study would be necessary before use of these materials could be assured.

MOLTEN SALTS IN LASER-INDUCED FUSION REACTORS

Two major uncertainties in use of Li_2BeF_4 in CTRs such as our hypothetical device stem from (a) potentailly troublesome interactions with the large magnetic field and (b) the fact that the first (plasma-containing) wall and blanket structure degrade the neutron spectrum so that the $^7Li(n,\alpha n')T$ reaction is reduced and tritium breeding becomes marginal. It is, accordingly, a matter of some interest to examine briefly the potential of molten salts in a CTR device which possesses neither a magnetic field nor a first wall.

Lubin and Fraas (33) have described a device in which pellets of deuterium and tritium produce a plasma upon ignition by an energy pulse from a suitable laser. The blanket-coolant fluid (Lubin and Fraas proposed metallic lithium) is pumped through the reaction vessel and through external power generation equipment. The liquid is pumped into the (essentially spherical) reaction vessel tangentially to provide a very rapid swirl; ignition of the pellet occurs in the vortex so formed on the vertical center-line of the vessel. The plasma generated in this pulsed device requires no magnetic containment, and the blanket coolant liquid is exposed directly to radiation from the plasma.

In addition to its coolant and breeding functions such a liquid must also provide attenuation of the severe shock waves sufficient to assure feasibility of the reactor vessel. Lubin and Fraas proposed to assist this function by introduction of (compressible) gas bubbles into the swirling liquid.

These shock waves are caused by (a) partial conversion of neutronic energy into mechanical energy within the liquid, and (b) deposition of x-ray energy in the liquid at the vortex surface. The former is by far

the dominant perturbing force (34). According to an analysis by Dresner (34), the impulse to the vessel wall due to the neutronically induced shock is proportional to

$$\alpha\mu C_p^{-1} R_1^{-1}$$

where α is the volume expansivity, μ is the sonic velocity, C_p is the specific heat, and R_1 is a neutron attenuation distance which depends upon neutron scattering and absorption reactions. The values of these four quantities for Li_2BeF_4 and for Li are:

	Li_2BeF_4 (600°C)	Li (600°C)
α (°C^{-1})	2.4×10^{-4} (11)	2.1×10^{-4} (35)
μ (cm sec^{-1})	3.0×10^{5a}	4.3×10^5 (36)
C_p (erg g^{-1} °C^{-1})	2.4×10^7 (11)	4.2×10^7 (35)
R_1 (cm)	23 (34)	33 (34)

[a]Estimated.

This analysis suggests that the walls of a vessel filled with molten Li_2BeF_4 will suffer an impulse almost twice that of an identical vessel filled with lithium metal. This conclusion, coupled with the fact that enhanced centrifugal forces (Li_2BeF_4 is nearly four times as dense as Li) will make suspension of gas bubbles more difficult in the salt, would seem to place Li_2BeF_4 at some disadvantage.[12]

It seems apparent, however, that design and construction of this vessel to withstand repeated shocks over a long life will pose formidable problems. Metallic lithium at temperatures of 500°C and above is likely to prove compatible with relatively few (and generally expensive and exotic) materials. It is possible that Li_2BeF_4 (or other salts), which are compatible with a much wider spectrum of metals may have real advantages in easing this difficult design problem.

Problems with chemical transmutations and with recovery and management of tritium seem generally similar to, and should be handled by methods like, those described above for the hypothetical toroidal device. It

[12]Differences in liquid properties will probably be relatively unimportant in attenuating the shock waves. The entrained bubbles almost certainly will be the principal shock-absorbers.

seems likely that the ease of tritium recovery may give the molten salts
an additional advantage.

Finally, it should be noted that the absence of the first wall leads
to decidedly improved breeding. Metallic lithium will still prove to
possess the highest ratios, but it seems certain that Li_2BeF_4 will have
values markedly above unity. Indeed, it is likely that several lithium-
bearing salt compositions would be possible breeders in this sort of
laser-powered CTR.

SUMMARY: GENERAL COMPARISON OF MOLTEN SALTS WITH LITHIUM IN FUSION REACTORS

In summary, and to supplement the several preceding discussions, we
briefly compare liquid lithium with salts (especially Li_2BeF_4) in several
regards.

Lithium metal is clearly superior to any molten salt in breeding of
tritium; this seems certainly true in any CTR embodiment. Certain pro-
posed designs, of which the laser-powered devices are the best examples,
can certainly breed sufficient tritium using molten salts alone. However,
the indication that tritium breeding is marginal for the salts in some
(if not most) designs represents the worst drawback to their use. Fluo-
ride salts are also inferior to lithium in that, primarily because of
fluorine's relatively high cross-section for ineleastic neutron scatter-
ing, the salts are more intense gamma sources and cause increased gamma
heating of the vacuum wall (12).

Tritium recovery should prove considerably simpler if molten salts
are used; this is particularly true if the tritium can be maintained as
TF to minimize diffusion through metallic walls.

Several of the physical properties of lithium (thermal conductivity,
specific heat, viscosity and melting point, for example) are superior to
those of the molten salt. However, since the magnetic field will prevent
turbulent flow in the blanket for lithium, but not for the salt, it may
be that the molten salt is a better heat transfer medium for such a CTR.

Lithium is compatible with relatively few structural metals, niobium
alloyed with 1% zirconium appears to be a suitable container (35). More-
over, lithium reacts with graphite, and this material must certainly be

clad if it is to serve in a lithium-cooled blanket assembly. Molten Li_2BeF_4 is compatible with graphite and with a wide variety of structural metals. Corrosion by the salt is possible, through interaction with strong magnetic fields, but such corrosion can apparently be avoided by careful design. Transmutations within the salt, which provide potential for corrosive reactions, can be accommodated by relatively simple means.

Reactions of salts with steam or with air produce accelerated corrosion but, unlike similar reactions of lithium, lead to no inherently hazardous conditions.

It is clearly not possible at this stage of the technology to predict with confidence how the problems inherent in use of molten salts (or of lithium) will be solved. It is entirely possible that both lithium and molten salts will be useful. At any event, and regardless of the ultimate choice, it is clear that many fascinating chemical research and development ventures lie ahead.

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