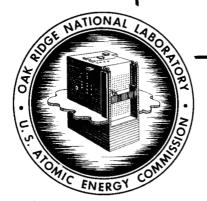


ORNL**-3258** UC-20 – Controlled Th**ermonuclear Processes** TID-4500 (17th ed.)

BLANKETS FOR THERMONUCLEAR REACTORS

C. J. Barton R. A. Strehlow

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REACTOR CHEMISTRY DIVISION

BLANKETS FOR THERMONUCLEAR REACTORS

C. J. Barton and R. A. Strehlow

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OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee operated by UNION CARBIDE CORPORATION for the U.S. ATOMIC ENERGY COMMISSION



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ABSTRACT

A study of blanket requirements for thermonuclear power reactors is presented and complemented with a short critical review of some blanket systems which have been suggested. An indication is made of some research problems associated with the blanket system presently considered most promising.

-iii-

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I. INTRODUCTION

Achievement of controlled thermonuclear power requires the solution of the very difficult problem of confining a stable energetic plasma. Concentration of research effort on this problem has resulted in little consideration of questions relating to energy extraction from a stable contained plasma. Many uncertainties exist regarding the design of a successful thermonuclear reactor but the fact remains that some fraction of the energy of thermonuclear neutrons must be removed or recovered as heat. Although this consideration alone makes necessary the presence of a heat recovery or removal blanket surrounding the reactor, the production of tritium is an equally necessary function of the blanket for a deuterium-tritium fueled (D-T) reactor. It is also essential that the highly energetic neutrons be prevented from damaging magnetic coil materials such as copper or sodium. The utilization of low-temperature superconducting coils will not, in itself, reduce the need for shielding. This report contains a brief discussion of thermonuclear reactions from the standpoint of energy recovery, a review of various blanket systems which have been suggested for use with thermonuclear reactors, some comments on problems connected with each type of blanket, and some suggestions for research on the blanket system presently considered most promising.

II. THERMONUCLEAR AND RELATED NUCLEAR REACTIONS

A confined thermonculear plasma will yield energy in the center of mass system in accordance with the following equations:¹

 $_{1}D^{2} + _{1}D^{2} \rightarrow _{2}He^{3}$ (0.82 Mev) + $_{0}n^{1}$ (2.45 Mev) (1)

$$_{1}D^{2} + _{1}D^{2} \rightarrow _{1}T^{3}$$
 (1.01 Mev) $+ _{1}H^{1}$ (3.02 Mev) (2)

$$_{1}D^{2} + _{1}T^{3} \rightarrow _{2}He^{4}$$
 (3.5 Mev) $+ _{0}n^{1}$ (14.1 Mev) (3)

$$_{1}D^{2} + _{2}He^{3} \rightarrow _{2}He^{4}$$
 (3.8 Mev) $+ _{1}H^{1}$ (14.7 Mev) (4)

The reactions in a deuterium-fueled (D-D) reactor can be described approximately by the sum of Equation (1) through (4) above, or by Equation (5) if the intermediate $_{2}\text{He}^{3}$ can be contained in the plasma long enough for the He³ (d,p) He⁴ reaction to take place.

 $3_{1}D^{2} \rightarrow {}_{2}\text{He}^{4}$ (3.6 Mev) + ${}_{0}n^{1}$ ($\frac{1}{2} \times 14.1 + \frac{1}{2} \times 2.95$) + ${}_{1}\text{H}^{1}$ (8.8 Mev) (5) The difference of 0.9 Mev between 21.6 Mev and the sum of these figures is the kinetic energy of the intermediate ${}_{1}T^{3}$ and ${}_{2}\text{He}^{3}$ nuclei. The deuterium-tritium (D-T) reaction is described by Equation (3) alone, and it is apparent that 80 percent of the total energy released in this type of thermonuclear reactor accompanies the neutrons. In a D-D reactor from 34 to 50 percent of the total energy released will be in the form of energetic neutrons depending on the fraction of He³ which reacts according to Equation (4).

Since the fraction of the total energy released in a D-D reactor which is associated with charged particles will be much larger than for a D-T reactor, there is a greater incentive to exploit some as yet undemonstrated method for directly converting the energy contained in a D-D reactor plasma into electrical power. While it is obviously desirable to recover the neutron energy in a D-D reactor, it is clear that development of a method for recovering the neutron energy is essential for the success of a D-T reactor.

Reactions (1) and (2) occur at approximately equal rates over a wide range of deuterium energies while the deuterium-tritium reaction (3)

occurs at a much faster rate below $\overline{E} \cong 600$ kev at all energy values. The D-He³ reaction (4) is slower than the other three at low energy levels but its rate equals that of the D-D reactions at about 120 kev. The technological problems associated with the attainment of a successful D-T reactor appear distinctly less formidable than for a D-D reactor. The larger part of the subsequent discussions which make up the body of this report is devoted to consideration of blanket materials for a D-T reactor while the requirements for a blanket to be employed with a D-D reactor are discussed briefly in the appendix. Some requirements for blanket materials are, of course, common to both types of reactors.

The neutrons liberated by the D-T reaction, after they have been slowed down to recover most of their energy, must be employed as efficiently as possible to produce the tritium needed to provide fuel for the reactor. The most obvious method for doing this is by reaction with Li⁶, which makes up 7.5 percent of naturally occurring lithium, according to the equation:

$$_{3}\text{Li}^{6} + _{0}\text{n}^{1} \rightarrow _{2}\text{He}^{4} + _{1}\text{T}^{3} + 4.6 \text{ Mev}$$
 (6)

If the energy released by this reaction is recovered as heat in the blanket, this would raise the amount of recoverable energy to 85 percent of the total released by each D-T reaction.

An alternate but less promising method of producing tritium is by reaction of neutrons with $B^{1\,0}$ according to the equation:

 ${}_{5}\mathbb{B}^{1\ 0} + {}_{0}n^{1} \rightarrow 2 {}_{2}\mathbb{H}e^{4} + {}_{1}\mathbb{T}^{3}.$ (7)

Some neutron losses are considered inevitable and it is, therefore, necessary to compensate for these losses by use of n,2n reactions. A number of these reactions (with lead, tungsten, or other materials with

-3-

Z greater than 40 or 50) are worthy of consideration but the only material that has been discussed to any extent in the literature on this subject is beryllium. The nuclear reaction involved in tritium production from beryllium is described by:

 $_{4}\text{Be}^{9} + _{0}\text{n}^{1} \rightarrow 2_{0}\text{n}^{1} + _{4}\text{Be}^{8} \rightarrow 2_{2}\text{He}^{4} + 2_{0}\text{n}^{1}$ (8) An alternate reaction of neutrons with beryllium results in the production of Li⁶ which will in turn produce tritium, but at a cost of 2 neutrons per atom of tritium produced. Fortunately, the cross section for reaction (8) is much larger than for the reaction producing either Li⁶ or Be¹⁰. Further discussion of neutron multipliers is found in Section V.

III. BLANKET REQUIREMENTS FOR A D-T REACTOR

Since design details of a successful thermonuclear power device are not known, it appears advisable to consider only the two principal functions which a blanket assembly will be called upon to perform and to discuss factors which are pertinent to the fulfillment of these functions. Fig. 1 from Rose and Clark² displays the features needed in a blanket assembly required to moderate and multiply 14.1 Mev neutrons from a D-T plasma as well as to breed tritium. These functions can probably be most readily performed by fluid blanket components which will allow heat extraction at reasonably high temperatures. Some factors that must be considered in choosing a suitable breeding and heat removal blanket are:

1. Provision for neutron multiplication

2. Moderation of neutrons to low energy and their reaction with 3Li⁶

-4-

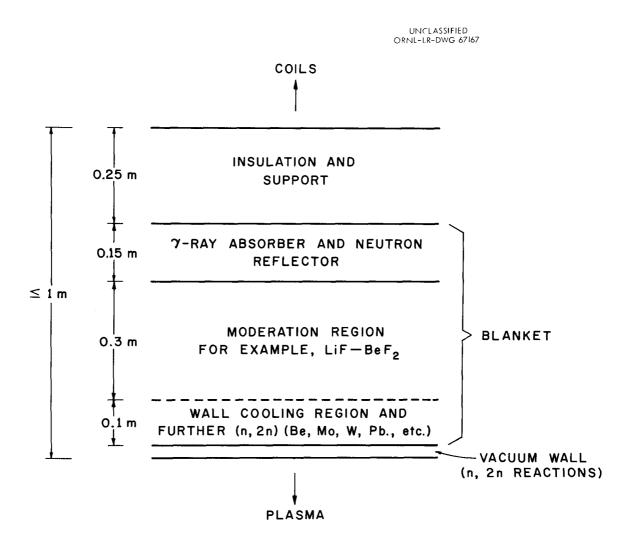


Fig. 1. Schematic Blanket Design for a Thermonuclear Reactor.

- -6-
- 3. Small parasitic neutron capture
- 4. Absorption of secondary γ -rays
- 5. Extraction of heat at useful temperatures
- 6. Recovery of tritium
- 7. Stability and chemical compatibility
- 8. Compatibility of the blanket with magnetic requirements, small electrical loss
- 9. The pressure of the system which should be low at the operating temperature
- 10. Minimum induced radioactivity
- 11. Minimum thickness

IV. BREEDING BLANKETS PREVIOUSLY SUGGESTED

Several blanket assemblies have been suggested which utilize a variety of chemical systems. The order in which these are considered below bears no relationship to the value attached to the different systems.

A. Solid Blanket Materials

Solids will be present in blanket systems for containment and possibly also for the purpose of providing moderation. Solid mixtures of Li_20 and BeO are mentioned by Robinson³ as a blanket material offering the possibility of breeding tritium and of neutron multiplication. The system was not studied in detail but there is noted the fact that LiOT and O_2 would be formed as chemically active corrosive materials when tritium is produced. Maintenance of material integrity would present a serious problem. Grimes⁴ states that tritium production could be accomplished in canned solid LiAl or other lithium alloy bathed in a liquid coolant. The problems mentioned for this type of assemblage are difficulty in fabricating the blanket and of providing suitable means for cooling. Use of water as the coolant would lead to either low grade steam or undesirably high pressures and would perhaps necessitate too great a blanket thickness. The use of sodium metal in addition to introducing high ohmic loss would also require the incorporation of additional moderating material. Grimes also mentions the possible use of a separately canned hydride moderator or a heavy metal reflector to partially degrade the neutron spectrum.

All solid blanket systems would require either a liquid or gaseous heat transfer medium. Periodic removal of the solid material and processing to recover the tritium would be required. Since it would be necessary to circulate a fluid through the blanket region, there appears to be no great incentive for separating the breeding and heat transfer tasks in this manner.

B. Slurries

The possibility of using Li_20 slurries in liquid LiNO_3 or LiNO_2 is considered by Johnson.⁵ It is his belief that due to the similarities in densities and magnetic properties, such slurried mixtures should be capable of being readily handled. However, Gruen⁶ points out that such a system is thermodynamically unstable in the presence of nitrogen oxides which would certainly be present due to thermal or radiation decomposition of the nitrogen containing species. Some of the problems specified or implied in Johnson's discussion are: solubility of tritoxide in fused

-7-

nitrate or nitrite as a function of temperature, extent of occlusion of tritoxide in Li_20 particles, and recovery of T_20 from a gaseous mixture containing oxygen and nitrogen oxides. A concentric double tube system is envisioned utilizing highly compressed water vapor in one set of tubes serving as moderator while the slurry would be pumped through the other set. The opinion is also stated that oxide-nitrate slurries could be contained in a variety of metals and that thermal decomposition of LiNO_3 would not be disadvantageous since the decomposition product, LiNO_2 , is stable to above 700°C. This opinion appears to be based upon unpublished information.

C. Fused LiNO3 and LiNO2

Johnson⁵ and Gruen⁶ discuss employment of liquid LiNO₃ or LiNO₂ and mixtures thereof as blanket materials. Molten LiNO₂ is also mentioned by Christofilos et al. in connection with an engineering study of the Astron thermonuclear reactor.⁷ A cost estimate of a tritium recovery system is presented by Johnson. The tritium produced in the blanket would be present in a gaseous mixture of tritium oxide, oxygen, and nitrogen oxides from which the tritium could be recovered. It would be necessary to have the concentration of nitrogen oxides relative to that of T₂O fairly large in order to repress the formation of LiOT in the fused salt which would complicate tritium recovery. Gruen points out that LiNO₃ is the thermodynamically stable phase in the presence of appreciable concentration of NO or NO₂. He states that no studies of radiation damage in fused nitrates or nitrites have been reported but that extensive decomposition will occur when they are subjected to radiation in a thermonuclear reactor. This belief is given support by studies

-8-

of radiation decomposition of aqueous nitrate solutions.^{8,9} The nature and extent of decomposition is difficult to predict since decomposition will result not only from collisions with neutrons but also from helium and tritium atoms produced as shown by equation (6) above. The assumption is made by Gruen that chemical equilibrium would be attained quickly thus tending to reverse the effects of radiation decomposition. However, Gruen further states his belief that the rate controlling step will be the diffusion of nitrogen oxides into the melt. This does not appear to be compatible with the assumption of a rapidly attained equilibrium.

It is clear that questions of this type can be answered only with extreme difficulty since suitably high fluxes of 14 Mev neutrons are not now readily available. Some of the assumptions made in Johnson's report (e.g., the possibility of recovering 99.999% of the tritium from the off-gas) would require processing capabilities which have not been demonstrated. A blanket system based upon fused nitrates or nitrites would require extensive experimentation to demonstrate its feasibility.

D. Metallic Lithium

Use of liquid lithium as a blanket material is discussed by Spitzer et al.,¹⁰ Johnson,¹¹ and Hinterman and Wideroe.¹² The analysis by Spitzer et al. is a theoretical study of a two tube system with one set of steel tubes containing water and one set containing lithium. The blanket composition by volume is Li 28.1%, H₂O 28.1%, and Fe 43.8%. Their calculations show that such a complete blanket must be 48 cm thick to reduce the neutron loss to 1%. They also point out that one of the principal obstacles in the use of metallic lithium as a blanket is the large amount of power required to pump it in a strong magnetic field.

-9-

Tritium recovery could be effected, according to Johnson,¹¹ by cooling the lithium to a temperature near its melting point, filtering off the precipitated LiT and heating the LiT to its decomposition temperature, or by a distillation or degassing process. Spitzer et al., also calculated the effect of adding 5% of beryllium by volume to the above mentioned blanket and concluded that 6.7% of the incident neutrons are absorbed by the Be⁹ (n,2n) reaction, so that this amount of neutrons will be available to compensate for other losses.

Several methods have been discussed for dealing with the problem of the large amount of power required to pump lithium in a strong magnetic field but none appears to be very satisfactory. Johnson¹¹ mentions the possibility of lining the lithium-containing tubes with a non-conducting material but does not discuss this concept. Spitzer et al. also mention this possibility and they state that a nonconducting coating which will not corrode nor crack and which can be applied to steel remains to be found. The requirement that the insulating material must withstand corrosion by hot lithium in addition to thermal stresses poses serious technical problems. Spitzer et al. also discuss the possibility of allowing the lithium to remain stagnant and effecting heat removal from the blanket by means of the water moderator. The lithium would presumably be heated to a rather high temperature, possibly greater than 1000° C. The facts that severe thermal stresses would result and that pressurized water would probably be required do not make this solution of the pumping problem particularly attractive. Lithium has also been suggested by $Creutz^{1,3}$ for thermonuclear power and by Clauser and Weibel^{1,4} as the working fluid in a thermonuclear rocket propulsion device.

-10-

E. LiF-BeF₂

The possibility of using molten LiF-BeF_2 has been suggested by Grimes,⁴ Davis,¹⁵ and Robinson³ and by Rose and Clark.² Some of the advantages of this fluid are:

- 1. Well established phase relationships and well-known physical properties (e.g., density and viscosity) exist
- 2. Demonstrated easy containment (in graphite or INOR-8)
- 3. Thermodynamic stability and low vapor pressures
- 4. Probable absence of serious radiation damage
- 5. The presence of neutron multiplying and moderating species having small parasitic neutron capture
- The molar ratio of lithium to beryllium can be varied from about 1:1 to 2:1
- 7. Extensive experience in the engineering aspects of handling and purifying molten LiF-BeF₂ has been obtained at ORNL. Pumps, heat exchangers, flow meters, etc., needed to circulate and remove heat from large volumes of this type of material have been operated.

Estimates made by Rose^{16} indicate that a blanket containing only LiF-BeF₂ and structural material would be thicker than is desired. By adding a region specifically designed to moderate and multiply neutrons, a neutron spectrum and flux might be achieved which would meet the stringent blanket thickness requirement somewhat better. This possibility is discussed below. The solubility of tritium and tritium fluoride in mixtures of LiF and BeF₂ is not known but estimates can be made based on existing data. Lack of detailed estimates of possible neutron fluxes impedes consideration of the problem of corrosion of container materials due to the charge imbalance occasioned by the $\text{Li}^6(n,\alpha)T^3$ reaction which is of an oxidizing nature,³ but Grimes¹⁷ indicates that a solution to this problem should not be hard to find. Although theoretical studies which may indicate the relative importance of competing nuclear reactions and optimum isotopic and chemical composition have been undertaken by Rose, it is not yet possible to state with precision the values of any parameters applicable to the LiF-BeF₂ system.

It does appear, however, that despite the lack of much information needed to complete the evaluation of molten LiF-BeF₂ as a blanket material, this fluid meets more of the requirements listed in Section III above than does any other fluid considered to date. With a suitable γ -ray shield all of the requirements listed are met with ease. Furthermore, the existence of a demonstrated technological capability in purification and handling of materials of this type serves to make it even more attractive.

V. NEUTRON MULTIPLYING BLANKETS

As mentioned in Section III, Rose and Clark² have considered a two region blanket assembly. The inner region serves to cool the vacuum wall as well as to allow neutron multiplication. The outer region employed for neutron moderation and tritium production would be of relatively simple design. Of course, these two regions can be interspersed and co-mingled to allow a smaller thermal load to exist at the vacuum wall. This co-mingling was suggested also by E. P. Blizard¹⁸ and R. E. Clausing¹⁹ who considered the moderation of 14.1 Mev neutrons by inelastic scattering

-12-

in heavy metals but did not discuss the (n,2n) reaction and the consequent multiplying function these metals would perform.

Fig. 2 from Rose and $Clark^2$ shows the (n,2n) cross sections for a variety of elements as well as fast fission cross sections for uranium-238. The possible utilization of uranium which introduces the problem of high radioactivity levels is at first glance unappealing. The high (n,2n) cross-section values for the heavy metals as compared with those for beryllium might allow a more rapid moderation for the most energetic neutrons and the required blanket thickness may be significantly less if a heavy metal is substituted for beryllium.

Consideration of the fact that quantities of the neutron multiplier material will be consumed in a D-T machine (as Li⁶ will also be) leads to the hope that one of the more plentiful elements might be used for this purpose. Inasmuch as the multiplying blanket is subject to most of the requirements discussed above in Section III, a stable molten salt may be considered for this application. Neglecting the vacuum wall problem temporarily, elements as light as zirconium may be considered. Possible fluids must meet the requirements of radiation and thermal stability, of minimum induced radioactivity, of usefully low melting points, and of reasonable availability. Mixtures of PbF_2 , ZrF_4 , BeF_2 , or SnF_2 with one or more alkali metal halides appear to be potentially useful in this regard. Although the ease with which SnF_2 and PbF_2 can be reduced may result in a severe containment problem for mixtures containing these materials, the advantages in their use, particularly for PbF_2 , indicate that they should not be eliminated from consideration without further study of this problem. Although an activation problem exists with iodine,

-13-

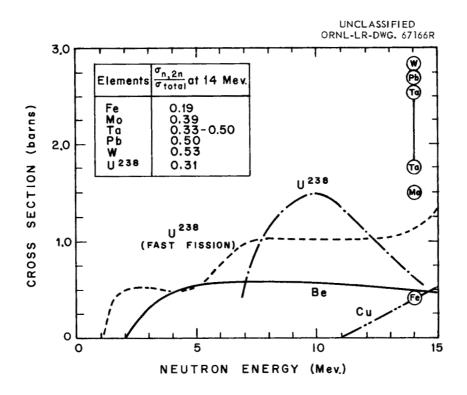


Fig. 2. Selected Neutron Multiplication Cross Sections.

cesium, and bismuth, incorporation of such components would allow further reduction of blanket thickness. A heterogeneous blanket assembly consisting of metallic tantalum, molybdenum, tungsten, etc., cooled by a suitable fluid would similarly be a reasonable choice.¹⁸ The fluid for the inner annuli of a cylindrically symmetrical reactor would not necessarily contain lithium. The problems of compatibility as well as mechanical design have not been thoroughly examined for such an assemblage.

The fact that neutrons of great energy are produced in a D-T reaction and that such a large number of elements are available for multiplication of the neutrons means that it is possible to breed fissionable fuel as well as tritium in thermonuclear reactors based upon the tritiumdeuterium reaction. This indicates that further consideration of possible neutron multiplying blankets is justified.

VI. THE VACUUM WALL

In addition to serving the function of providing a vacuum container the vacuum wall may also have a significant effect on the neutrons that pass through it and thus may be considered to be an integral part of the blanket system. The part of the wall facing the plasma will probably be subjected to intense heating and a special cooling effort may be required to maintain this wall at a moderate temperature. Tungsten, molybdenum, tantalum, rhenium, hafnium, niobium, and graphite are among those materials which are conceivably useful as wall materials because of their low vapor pressures and other favorable properties. Most of these materials act as neutron multipliers and it may be necessary to be economical in their use in order to minimize consumption of relatively

-15-

rare elements. If a thermonuclear reactor is designed to operate profitably at low magnetic induction (1.5 webers/ m^2), the energy flux upon the wall would be expected to be moderate. The wall might be damaged by:

- 1. The fast neutron flux
- 2. Sputtering from the surface by fast plasma ions
- 3. Failure to remove the radiant heat incident upon it from the plasma.

Item (1) is a necessary consequence of operation and Item (2) is probably primarily a plasma problem. The radiant energy, Item (3), consists of cyclotron resonance radiation and bremsstrahlung x-rays. Both are absorbed in a small depth of almost any of the above listed elements and to a first approximation may be assumed to be deposited at the vacuum surface. Since this energy could be recovered as heat, the cooled portion of the wall should run at a usefully high temperature.*

Table 1 from Craston et al.²⁰ lists possible heat flux rates for a number of materials under arbitrarily chosen conditions. The heat flux under these conditions (wall thickness 1 cm and coolant temperature 400°C) is determined by the thermal conductivity of the material and the temperature of the vacuum boundary surface. The temperature, Θ_{max} , picked for each material is that which yields equal vaporization rates. Cranston et al. also discuss some other materials considerations for pulsed devices which are not being directly considered in this report. Several conclusions may be tentatively stated:

-16-

^{*} The authors have drawn quite heavily from Rose and Clark² for the considerations presented in this section.

- 2. If the wall must be made of a single material, the refractory metals appear to be the best choice.
- 3. If the thermal flux is several kw/cm^2 , the wall must be thin (≤ 1 cm), and consequently, the coolant must run at a relatively low pressure.
- 4. A sandwich type wall, arranged for low corrosion on the coolant side and low vapor pressure on the plasma side may prove to be desirable.

Element	Heat Flux Through $\Theta_{max} ^{O}C$ for a Constant Erosion Rate	Wall and Wall Thickness Watts/cm ² for l cm Thickness with Θ _{coolant} = 400 ^o C
Beryllium	670	300
Magnesium	140	-
Aluminum	510	240
Titanium	840	60
Vanadium	1080	160
Chromium	630	120
Cobalt	410	260
Nickel	830	260
Copper	580	640
Zirconium	1150	90
Molybdenum	1480	1080
Alumina	1030	40
Beryllia	1360	180
Silica	610	4

Table 1. Maximum Internal Wall Temperatures

VII. CONCLUSIONS

Existing information indicates that molten LiF-BeF_2 is a promising blanket material for removing energy from a thermonuclear reactor in the form of useful heat and for breeding tritium. It is also apparent that, in the interest of minimizing blanket thickness and of maximizing neutron multiplication, the possibility of including heavier elements such as lead, tin, barium, and zirconium in a fluoride salt mixture to be placed in one region of a blanket assembly should be considered. It is probably not too early to start to obtain information needed to determine the feasibility of employing molten fluorides in a thermonuclear reactor blanket. Some of the problems that need to be examined are: compatibility of molten fluorides with container and neutron multiplying materials and means of dealing with the corrosion problem resulting from charge imbalance accompanying tritium production, and solubility of tritium and tritium fluoride in molten LiF-BeF2. Due to lack of information on the configuration of a successful thermonuclear reactor, it seems obvious that it would not be profitable to attempt blanket design studies at the present time.

-18-

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APPENDIX

Blanket Requirements for a Deuterium-Fueled Reactor

If a cyclotron radiation loss proves not to be excessively large, there is a strong possibility that a D-D reactor may be more nearly feasible than is implied in the body of this report. The blanket requirements for a thermonuclear reactor of this type listed below are different in several respects from those listed in Section III.

- 1. Neutrons must be moderated to low energy and absorbed.
- 2. Heat should be extracted at usefully high temperatures.
- 3. There should be a minimum of induced radioactivity, although parasitic capture of neutrons is not in itself considered to be a serious problem.
- 4. Chemical stability and compatibility are of the same order of importance as for the D-T blanket.
- 5. A much more stringent magnetic requirement may be imposed, as compared to that for a D-T blanket.
- 6. A low pressure system is more desirable.
- 7. Minimum thickness is probably a necessary requirement.

The principal differences between this list and the one in Section III of the present report are: first, no immediate concern with neutron economy; second, a probably more stringent magnetic compatibility requirement; and third, the problem of what to do with the neutrons mentioned in Item (1). One solution of this problem would be to produce fissionable material by absorption in fertile materials such as U^{238} or Th^{232} . The neutrons could presumably be used to produce various useful isotopes but it is possible only to speculate at present concerning the market for such materials.

In a D-D reactor there will be approximately equal numbers of 14 and 2.5 Mev neutrons which must be moderated. Molten salt or metal would again be a reasonable choice as heat transfer agent. Either type of material could extract energy as heat at usefully high temperatures without an unduly thick assembly.

Since the blanket assembly may be subjected to varying magnetic fields in a D-D reactor, metals or good conductors might be usable if at all only in strictly limited amounts. One way in which these requirements could be met is by use of a heavy metal molten salt contained in a ceramic material having only metals of low atomic number. The metallic structure for such an assembly could be designed to prevent induced currents from diverting power. It can be seen that although there are differences in the requirements for D-D and D-T reactor blankets, there are also a number of requirements which are common to both types of reactors.

-22-

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