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**THE REQUIREMENTS FOR PROCESSING TRITIUM RECOVERED FROM
LIQUID LITHIUM BLANKETS:**

THE BLANKET INTERFACE*

R. G. Clemmer, P. A. Finn, L. R. Greenwood, T. L. Grimm, and D. K. Sze
Chemical Technology Division/Fusion Power Program
Argonne National Laboratory
9700 S. Cass Ave., Argonne, IL 60439 USA

and

J. R. Bartlit and J. L. Anderson
Tritium Systems Test Assembly
Los Alamos National Laboratory
P. O. Box 1663, Los Alamos, NM 87545 USA

MASTER

and

H. Yoshida and Y. Naruse
Japan Atomic Energy Research Institute
Shirakata, Tokai-mura, Nakagun, Ibaraki-ken 319-11 JAPAN

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TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
1. INTRODUCTION	3
2.1 Tritium Recovery Options	8
2.2 Permeation Window	12
2.2.1 Permeation/Oxidation	12
2.2.2 Permeation/Cold Trap	14
2.3 Molten Salt Recovery Process	15
3. EVALUATION OF TRITIUM RECOVERY METHODS	17
3.1 Extraction Methods - Evaluation	17
3.2 Reference-Method Selection	22
4. REFERENCE SYSTEM	24
5. EVALUATION OF TRANSPORT RATES OF IMPURITIES	28
5.1 Source Terms for Protium (H) in Lithium	28
5.1.1 Permeation of Protium from the Intermediate Sodium Loop	28
5.1.2 Injection of Deuterium and Hydrogen from the Plasma through the First Wall	29
5.1.3 Production of H from (n,p) Reactions in the Structure	29
5.1.4 H Residue in the Lithium	29
5.1.5 H Residue in the Structure	29
5.1.6 Miscellaneous	30
5.1.7 Total Source of H into Li	30
5.2 Other Lithium Contaminants - LMFBR and Lithium Loop Operation	31
5.3 Assessment of Amounts of Other Impurities in Lithium, Including Nonmetallic Elements, Metallic Elements, and Particulates	34

TABLE OF CONTENTS

(Continued)

	<u>Page</u>
5.3.1 Nonmetallic Elements in Lithium	34
5.3.2 Metallic Impurities in Lithium	36
5.3.3 Particulates in Lithium	38
5.4 Activity Levels in Lithium	38
6. TRANSPORT OF IMPURITIES IN THE MOLTEN SALT CYCLE	40
6.1 Transfer of Impurities from the Lithium to the Salt	40
6.2 Radionuclides in the Salt	41
6.3 Electrolysis	41
7. CONDITION OF THE SPARGE GAS STREAM	44
8. CONCLUSIONS	46
ACKNOWLEDGEMENTS	47
REFERENCES	48

LIST OF FIGURES

<u>Number</u>	<u>Title</u>	<u>Page</u>
1	Plateau pressures of the monohydrides of various metals	6
2	T ₂ pressure over dilute solutions of tritium in lithium	7
3	T ₂ pressures above yttrium	9
4	T ₂ pressures above yttrium for various T/Y ratios in the Sieverts' region	10
5	Equilibrium tritium (T ₂) pressures above yttrium (T/Y = 0.001) and lithium (1 wppm T)	11
6	Flow sheet for blanket tritium systems	26

LIST OF TABLES

<u>Number</u>	<u>Title</u>	<u>Page</u>
1	Parameters of a Permeation/Oxidation Tritium Recovery System	13
2	An Evaluation of Four Methods to Extract Tritium from Liquid Lithium	23
3	Reference Parameters for the Blanket Tritium System	26
4	Sources of H into Li	30
5	Observed Impurities in Operating LMFBRs	32
6	Observed Impurities in Operating Experimental Lithium Loops	33
7	Chemical Composition of HT-9 (Sandvik)	35
8	Typical Impurity Levels of Nonmetallic Elements in Liquid Metal Systems	37
9	Concentration of Dissolved Metallic Elements in Lithium	37
10	Activity Levels in Lithium	39
11	Transfer of Impurities from the Lithium into the Salt	42

LIST OF FIGURES
(Continued)

<u>Number</u>	<u>Title</u>	<u>Page</u>
12	Radionuclides in the Salt	43
13	Conditions of Sparge Gas	45

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ABSTRACT

The requirements of tritium technology are centered in three main areas, (1) fuel processing, (2) breeder tritium extraction, and (3) tritium containment. The Tritium Systems Test Assembly (TSTA) now in operation at Los Alamos National Laboratory (LANL) is dedicated to developing and demonstrating the tritium technology for fuel processing and containment. TSTA is the only fusion fuel processing facility that can operate in a continuous closed-loop mode. The tritium throughput of TSTA is 1000 g/d. However, TSTA does not have a blanket interface system. The initial TSTA proposal of 1976 included a blanket interface, as part of TSTA, to simulate the processing of the tritium gas stream from the blanket extraction system. However, due to a limited budget and limited information on blanket systems at that time, the blanket interface was not included.

Blanket processing is an important part of the fusion fuel cycle. It is the interface between the blanket tritium extraction system and the fuel processing system. Although the tritium throughput in the plasma exhaust is a factor of 20 to 100 higher than in the blanket system (corresponding to a plasma burn fraction of 1-5%), the total hydrogen isotope throughput may be dominated by the blanket stream. For the aqueous solution blanket, a candidate for both U.S. ITER and NET fusion reactor designs, the hydrogen throughput in the blanket system is a factor of 1000 higher than in the plasma exhaust. In addition, the helium content, impurity composition, and radionuclides in the blanket stream are very different from those in the plasma exhaust. Therefore, to develop and demonstrate tritium technology for fuel processing, it is important to add blanket processing to TSTA. The goal of the TSTA experiments is to demonstrate that the fusion fuel cycle can be operated under the constraints of engineering, environmental safety, and economics.

We have initiated a study to define a blanket processing mockup for TSTA. Initial evaluation of the requirements of the blanket processing system have been started. The first step of the work is to define the condition of the gaseous tritium stream from the blanket tritium recovery system. This report summarizes this part of the work for one particular blanket concept, i.e., a self-cooled lithium blanket. The total gas throughput, the hydrogen to tritium ratio, the corrosive chemicals, and the radionuclides are defined. The key discoveries are:

1. The throughput of the blanket gas stream (including the helium carrier gas) is about two orders of magnitude higher than the plasma exhaust stream.
2. The protium to tritium ratio is about 1. The deuterium to tritium ratio is about 0.003.
3. The corrosion chemicals are dominated by halides.
4. The radionuclides are dominated by C-14, P-32, and S-35.
5. There is high level of nitrogen contamination in the blanket stream.

Future work will be required to define the processing system to clean up this stream and its interface with TSTA. Similar work for the aqueous solution blanket and helium-cooled, Li_2O blanket will be conducted.

1. INTRODUCTION

The requirements of tritium technology are centered in three main areas, (1) fuel processing, (2) breeder tritium extraction, and (3) tritium containment. The Tritium Systems Test Assembly (TSTA) [1,2] now in operation at Los Alamos National Laboratory (LANL) is dedicated to developing and demonstrating the tritium technology for fuel processing and containment. TSTA is the only fusion fuel processing facility that can operate in a continuous closed-loop mode. The tritium throughput of TSTA is 1000 g/d. However, TSTA does not have a blanket interface system. The initial TSTA proposal of 1976 included a blanket interface, as part of TSTA, to simulate the processing of the tritium gas stream from the blanket extraction system. However, due to a limited budget and limited information on blanket systems at that time, the blanket interface was not included.

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TSTA (e.g., at the inlet of the Fuel Cleanup Unit [FCU] or the inlet of the Isotope Separation System [ISS], or possibly at other points in the TSTA system) will be evaluated. The specific goal is to identify which operations (purification, isotope enrichment, etc.) require special handling for the blanket streams.

In support of this effort, the characteristics of the blanket tritium streams from one type of blanket were evaluated: namely, self-cooled liquid lithium. Later phases of the study will investigate other blanket concepts, including Li_2O - a solid breeder blanket and an aqueous solution blanket. We assumed that the reference fusion reactor had the following parameters: thermal power of 1000 MW(t), thermal energy of 20 MeV per fusion, a lithium coolant temperature of 300°C at the inlet of the blanket and 500°C at the outlet, and a tritium breeding ratio of 1.10. To further define the blanket system, the method of tritium recovery was selected. Then the liquid lithium system was analyzed in sufficient detail to define the flow of impurities, etc., in the gaseous output tritium stream recovered from the blanket.

Reported herein are (1) an assessment of the options for tritium recovery from liquid lithium and the selection of the reference method (molten salt extraction); (2) a definition of reference blanket, salt, and sparge stream conditions; (3) an evaluation of the source terms in each stream including impurities; and (4) the determination of the condition of the gaseous blanket tritium stream.

2. TRITIUM RECOVERY OPTIONS

Various methods of tritium recovery from liquid lithium were assessed. Presented in this section are a description of lithium chemistry and descriptions of the methods of tritium recovery. The methods considered were (1) yttrium gettering, (2) permeation windows, and (3) molten salt extraction. Comparative evaluation and selection of the reference method of tritium recovery are presented in Section III. For the self-cooled liquid lithium blanket system, the reference reactor parameters are: a thermal power of 1000 MW; lithium inlet and outlet temperatures of 300°C and 500°C, respectively; and HT-9 structure. This reference reactor concept should be applicable in many respects to a blanket with vanadium alloy structure or to a blanket that has static lithium cooled with helium.

The chemistry of lithium is characterized by a strong tendency to react with nonmetallic elements, notably oxygen, nitrogen, and hydrogen [3]. The plateau pressures of hydrogen above lithium are rather low in comparison to most other metals, and are comparable to strong hydride formers like titanium, zirconium, and hafnium (see Fig. 1, from Vertrano [4]). The low plateau pressure for hydrogen above lithium-lithium hydride indicates that the decomposition pressure of the lithium hydride is low and, therefore, lithium hydride is quite stable. At lower concentrations of hydrogen isotopes in liquid lithium, the hydrogen (or tritium) is in solution and Sieverts' law applies [square root of $H_2(T_2)$ pressure is proportional to the concentration in solution]. Sieverts' constants for hydrogen isotopes in lithium have been measured [5-9]. The T_2 pressures over lithium for various concentrations as derived from the Sieverts' constants reported by Smith [8] are shown in Fig. 2. It can be seen that at 500°C, with 1 wppm tritium in lithium, the equilibrium pressure is only about 10^{-7} Pa. At lower temperatures, the pressure is even lower. Thus, lithium is quite a good getter for tritium. This means that recovery of tritium from liquid lithium to very low levels is difficult. There appear to be few methods that may be able to recover tritium from lithium that has a tritium content of 1 wppm: (1) yttrium gettering, (2) permeation windows, and (3) molten salt extraction.

Plateau Pressures of The Monohydrides(from Vertrano)

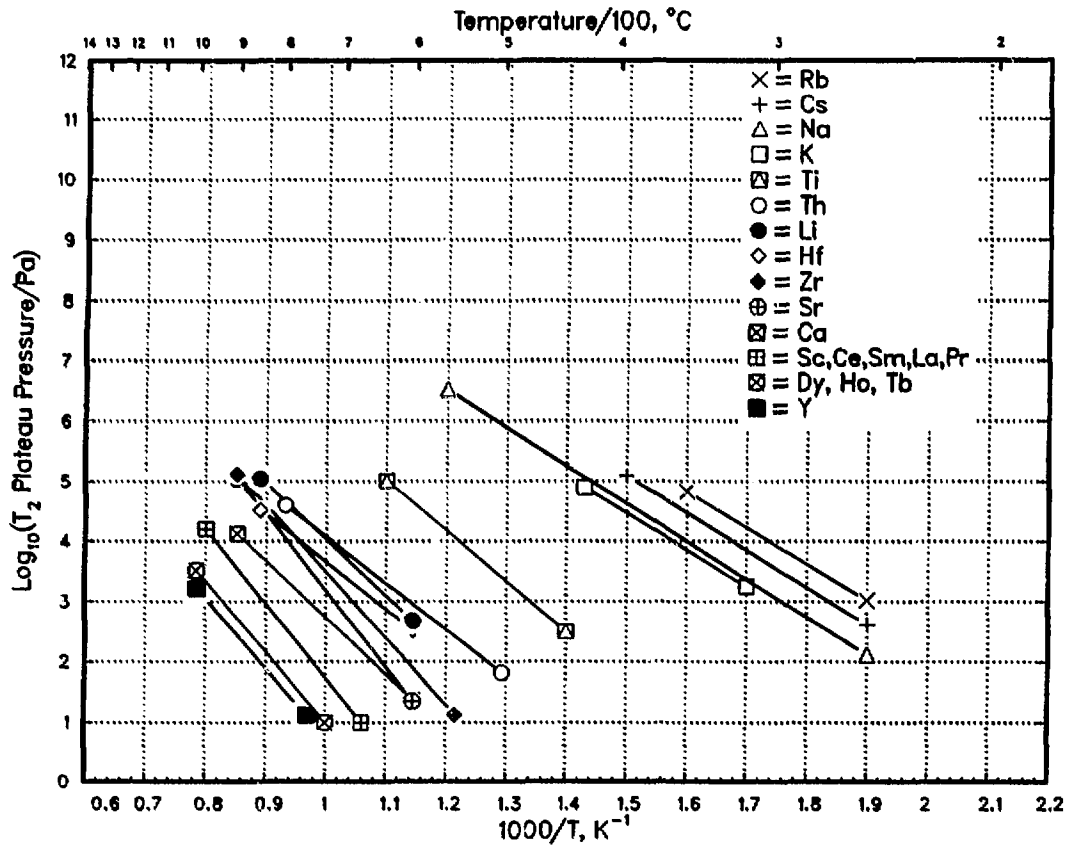


Figure 1 Plateau pressures of the monohydrides of various metals. (From Vertrano [4].)

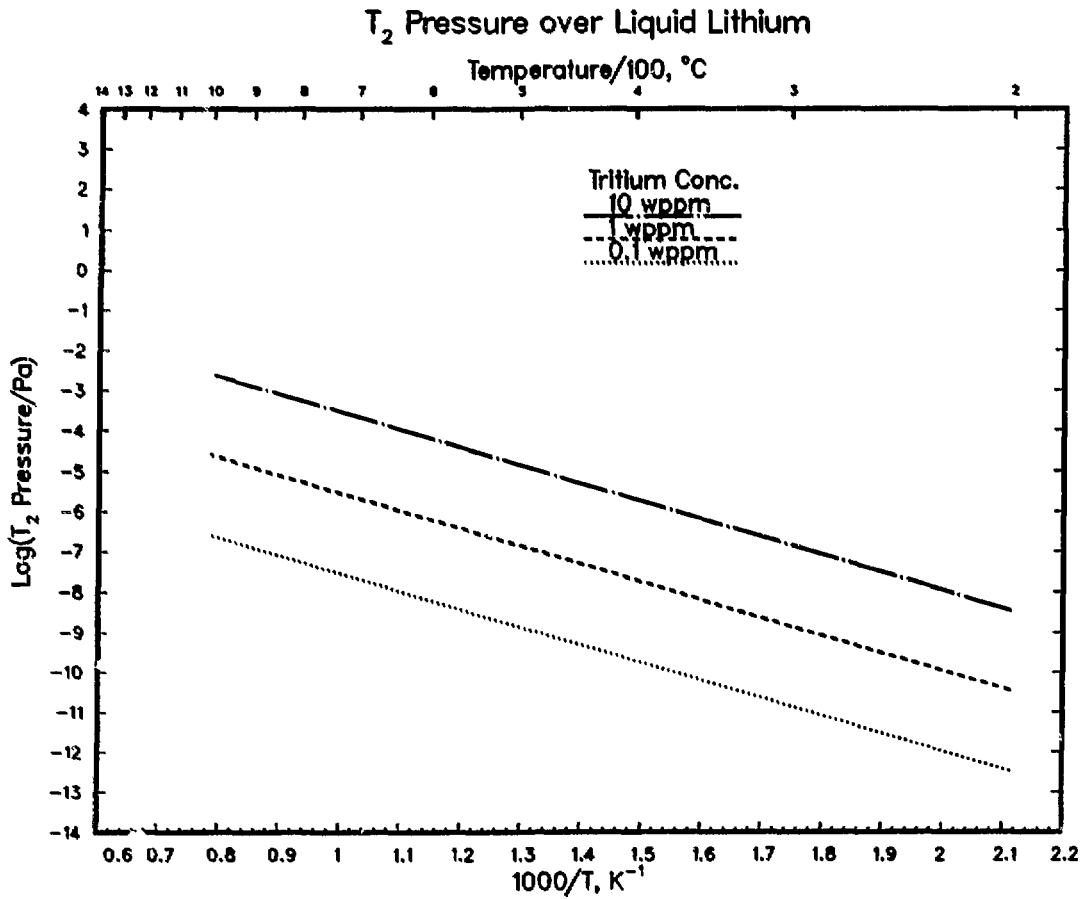


Figure 2 T_2 pressure over dilute solutions of tritium in lithium.

2.1 Yttrium Gettering

One potential method of extracting tritium from liquid lithium is by using a metal getter such as yttrium [10-21]. Referring to Figure 1, the material that forms the most stable hydride, thermodynamically the best getter, is yttrium. The getter could be used in the plateau region (T/Y ratio up to about 1) or possibly in the Sieverts' region (T/Y ratio 0.10 or less). Data from three studies [15,18,20] are plotted in Figure 3. The plateau pressures for tritium were based on data for hydrogen [15] and corrected for estimated isotope effects by multiplying the hydrogen pressures by a factor of $\sqrt{3}$. The Sieverts' data [18,20] were calculated for a T/Y ratio of 0.01. Also shown is the vapor pressure of T_2 above lithium with 1 wppm tritium. The best data set is that of the plateau pressures, from Yannopolis et al. [15]. When the plateau pressures are extrapolated to low temperatures, the plateau data appear to cross over the Sieverts' data. It is suspected that kinetic factors may be affecting the Sieverts' data. Also, the data of Clinton and Watson [18] are for very low concentrations (10-100 appm tritium in yttrium) and the extrapolation to a T/Y ratio of 0.01 (10^4 appm) is two to three orders of magnitude in concentration. Thus data have a considerable degree of uncertainty, particularly at low temperatures. This uncertainty is estimated to be less than one order of magnitude at 700°C and about two orders of magnitude at 300°C.

The equilibrium partial pressure of tritium above yttrium for T/Y ratios of 0.01, 0.001, and 0.0001 are illustrated in Figure 4, from the data of Clinton and Watson [18]. In Figure 5, the pressures for T/Y = 0.001 are compared to lithium with 1 wppm tritium. For the getter to function, it must have a lower pressure than that of the lithium. Thus, for the yttrium to getter 1 wppm tritium from lithium, the T/Y ratio at 300°C must be 0.001 or less, as shown.

Because the data are for equilibrium conditions only, the values are the best possible to do with yttrium gettering. Kinetic factors are likely to reduce the effectiveness. Even from the reference point in Figure 5, it is apparent that low temperatures (~ 300°C) and rather high Y-to-T ratios (~ 1000) are required. A specific problem with yttrium is that it is also an excellent oxygen scavenger, and an oxide film on yttrium has an extremely low tritium diffusivity, about 10^{-16} cm²/s at 500°C [19]. Further problems with

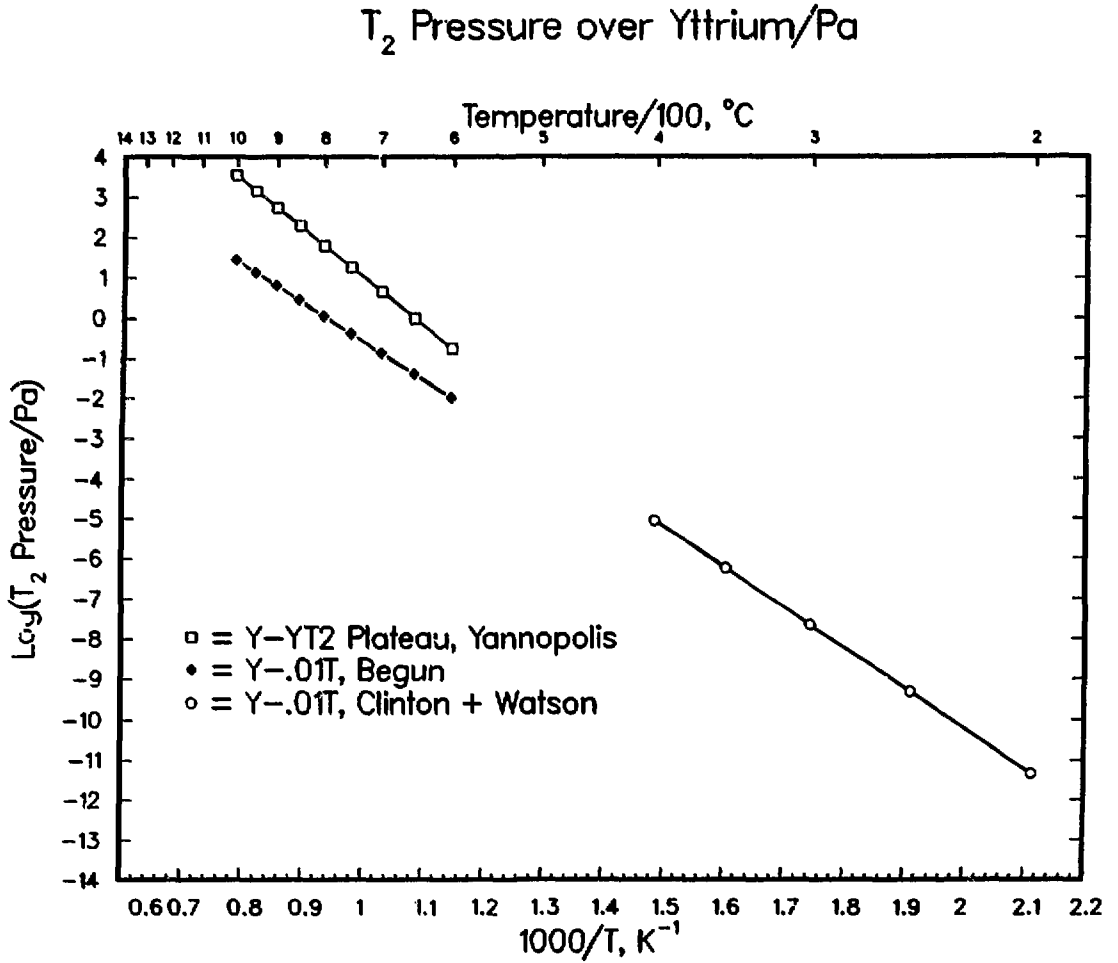


Figure 3 T₂ pressure above yttrium (from Refs. [15], [18], and [20]).

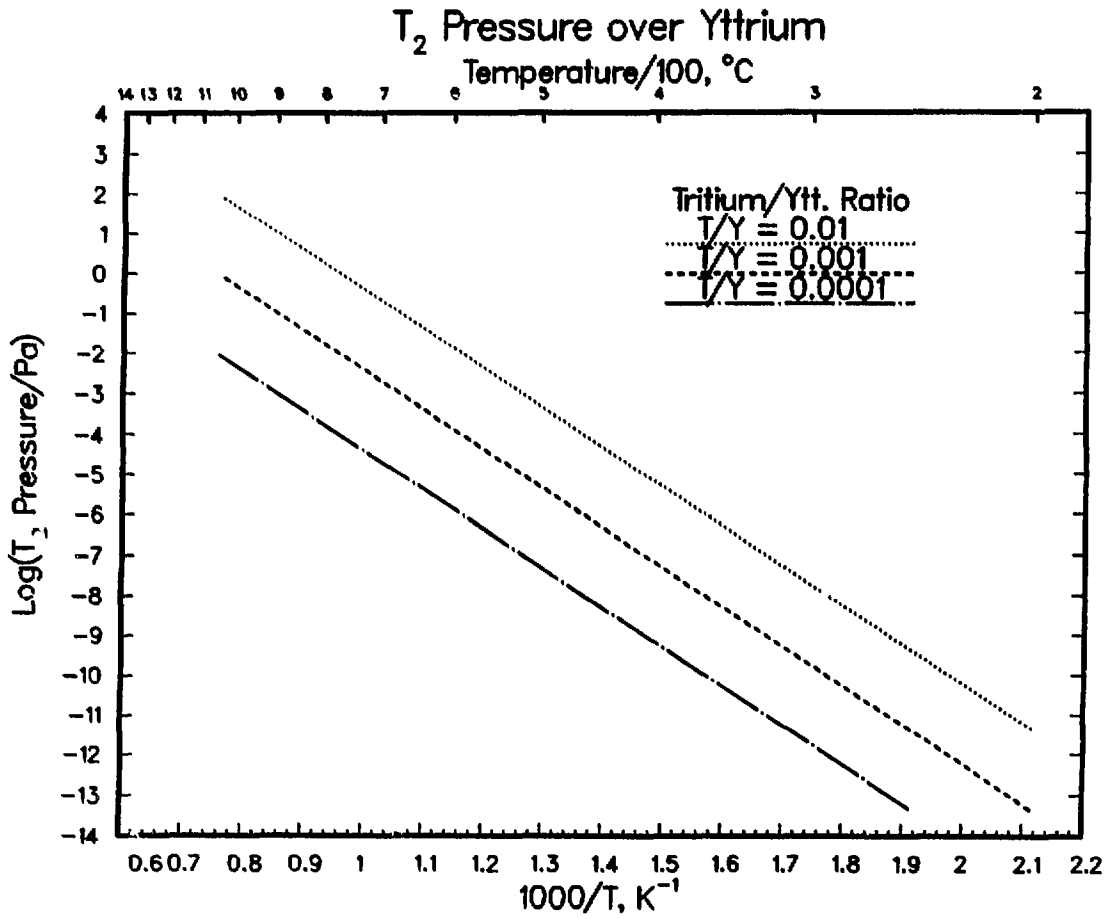


Figure 4 T_2 pressure above yttrium for various T/Y ratios in the Sievert's region.

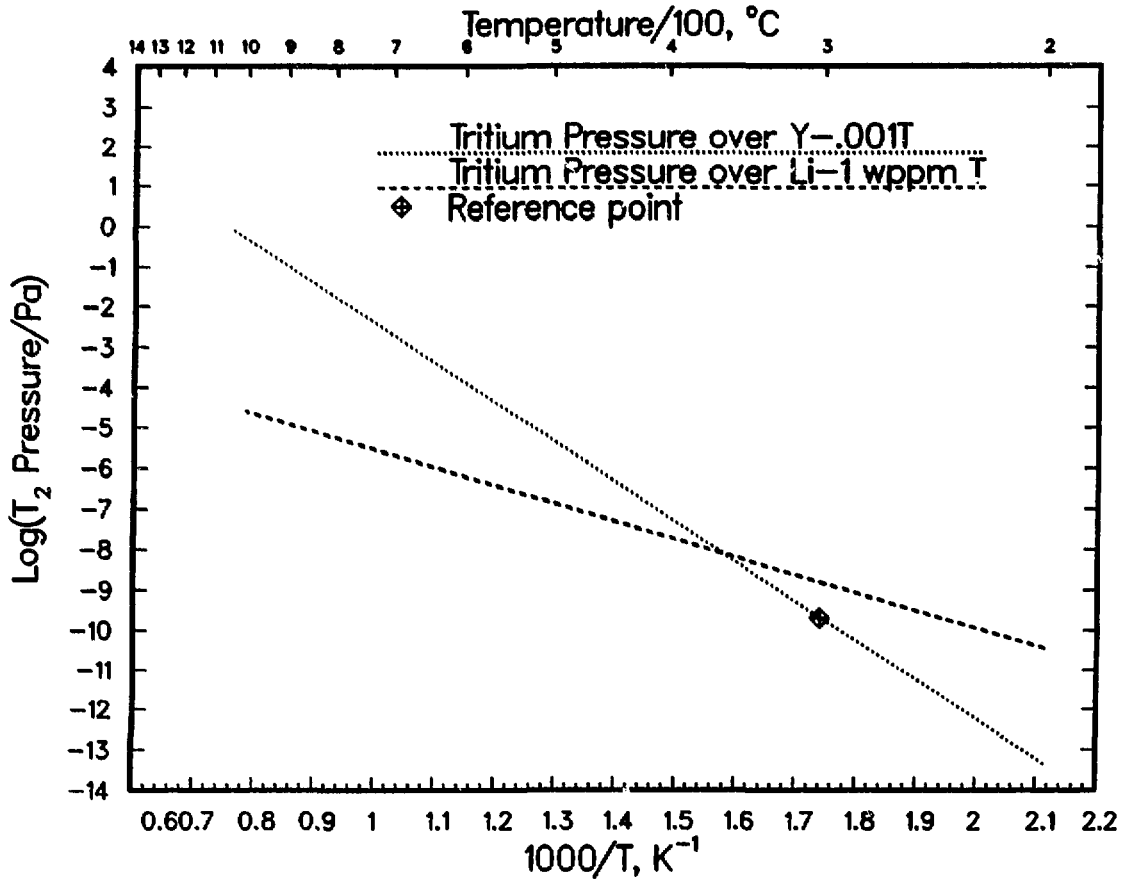


Figure 5 Equilibrium (T_2) pressures above yttrium ($T/Y = 0.001$) and lithium (1 wppm T).

yttrium are that it is very expensive as a result of its relative scarcity and is difficult to fabricate because of its tendency to form oxide. Small amounts of foil are estimated to cost \$10.00 to \$15.00 per square inch (Research Chemicals, Inc., Phoenix, Arizona). However, larger amounts will be much less costly estimated to be \$185 per pound for raw ingot (Molycorp). If it is assumed that the getter is on line for 12 hours and it takes 12 hours to regenerate the bed, then two beds are required. Each bed is to sorb 75 g of tritium, and each bed contains 2.2 metric tons of yttrium. The cost of 4.45 metric tons of yttrium at \$200 per pound is about \$2 million.

2.2 Permeation Windows

The permeation rate of hydrogen (tritium) through some metallic membranes can be very high, especially at high temperature [16,22-24]. This phenomenon is the basis for the permeation-window method for tritium recovery from lithium. The tritium still has to be removed from the downstream side of the permeation window. This step is difficult, mainly because of the very low driving force of tritium mass transfer from lithium. At 1 wppm tritium concentration, the tritium partial pressure in lithium is 10^{-7} Pa at 500°C. The tritium partial pressure on the downstream side of the window must be lower than this. The different methods proposed for efficient removal of tritium at such a low pressure constitute the main difference between the two permeation-window concepts, i.e., permeation/oxidation [24] and permeation/cold trap [25]. Those two different processes are discussed separately.

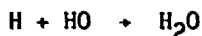
2.2.1 Permeation/Oxidation: The permeation window is made of zirconium with a palladium coating on the downstream side.[26] The zirconium is chosen for high tritium permeability and compatibility with lithium. The palladium coating is used to protect the zirconium from being oxidized and for its high tritium permeability and catalytic properties. An oxidizing atmosphere is maintained at the downstream side of the permeation window. The oxidation reaction on the surface of the palladium coating converts tritium (T_2) to tritiated water T_2O and, thus, retains the very low T_2 partial pressure that drives the diffusion [20]. The partial pressure of T_2O can be much higher than 10^{-7} Pa, and efficient removal by a purge gas becomes feasible.

The palladium diffuser used in TSTA [2] is based on the permeation-window concept [2]. The concept was demonstrated [2] in a scale considerably smaller than that required for a fusion reactor blanket. The tritium partial pressure was also much higher than is anticipated for a fusion reactor blanket. There are unique features in the blanket operation caused by the lithium environment, i.e., impurities transport and low tritium partial pressure. No experimental results are available for these conditions. As part of ANL's Tokamak Power Systems Studies (TPSS) effort [25], a tritium recovery system was designed based on the permeation/oxidation concept. The major parameters of the concept are summarized in Table 1. The costs are for materials only. The impact of a window with this large a surface area upon the overall system has yet to be investigated.

Table 1
Parameters of a Permeation/Oxidation Tritium Recovery System

Tritium recovery rate	150 g/d
Window temperature	500°C
Zr thickness	3.0 mm
Pa thickness	5.6 μm
Window area	$5.2 \times 10^3 \text{ m}^2$
Materials cost	\$7.4 million
System lifetime	30 years

In the calculation of the size of the permeation window, the possible effects caused by the impurity deposition on the window were not considered. If the impurity is in the form of an oxide, regeneration at higher temperature may be sufficient to decompose the oxide. If the impurity is in the form of carbide or corrosion product deposits, the window may have to be replaced periodically. The oxidation kinetics on the downstream side were assumed to be instantaneous; but, this assumption may be even more serious. It has been well established that the oxidation of hydrogen is first-order reaction at high hydrogen potential, but becomes to a second-order reaction at lower hydrogen potential, because the reaction is caused by



where the existence of HO is dominated by O at high hydrogen potential, but is dominated by H at lower hydrogen potential. Therefore, both the kinetics and thermodynamics become very unfavorable in the regime of interest. A possible method to alleviate this problem is to add hydrogen, as well as oxygen, in the purge gas. Then the reaction becomes



which becomes a first-order reaction for tritium. Thus kinetics and thermodynamics would be much more favorable for the release of tritium. (A similar effect has been observed for tritium release from solid breeders.) However, additional steps would be required to separate the added hydrogen from the tritium stream.

Permeation/oxidation is an interesting concept because of its simplicity. It can operate in a continuous process, so the tritium inventory can be low. It has yet to be demonstrated that the system can operate efficiently at the very low tritium partial pressure required for the fusion application. The key feasibility problems to be investigated are the thermodynamics and kinetics of oxidation on the interface between palladium and the purge gas. If significant amounts of hydrogen have to be added to enhance the oxidation process, the attractiveness of this concept will be significantly reduced. Because the chemistry of oxidation has not been demonstrated at the parameter regime in which we are interested, it is not possible at this time to define the purge-gas composition or determining the overall attractiveness of the concept. In addition, very large surface areas are required. The window unit(s) may be very large, costly, and have high lithium inventories. For these reasons, this concept is not selected for our work.

2.2.2 Permeation/Cold Trap: It has been demonstrated in the LMFBR [23-27] that tritium can be cold trapped and recovered from the sodium coolant [27]. At lower sodium temperature, NaT becomes super-saturated and forms particulates as its solubility decreases. The particulates can be recovered from the sodium by a filter system. Based on this principle, it was suggested [23] that tritium could be allowed to permeate from the primary lithium loop through the Intermediate Heat Exchanger (IHX) to the secondary sodium loop.

It is suggested that the material for the IHX be a vanadium alloy, because of its high permeability for tritium and good compatibility with lithium. A cold-trap system would then be used to recover the tritium from the sodium. However, a cold trap cannot reduce the tritium concentration to a corresponding partial pressure acceptable for fusion applications. Therefore, hydrogen would have to be added to the sodium so that, even with the total hydrogen partial pressure kept at 10^{-2} Pa, the tritium partial pressure could be kept to very low levels.

For tritium recovery from lithium, the tritium partial pressure in the lithium is 10^{-7} Pa at 500°C , which is also the maximum pressure allowable in the sodium. Because the cold trap can reduce the total hydrogen partial pressure only to 10^{-2} Pa, a H/T partial pressure of 10^5 must be maintained. Therefore, for each mole of tritium to be recovered, 10^5 moles of hydrogen must be added and separated from the tritium - a situation that is clearly not acceptable. Thus, it can be concluded that, for the temperatures considered herein, the permeation/cold trap concept, even though it was demonstrated to recover a very small amount of tritium from an LMFBR, cannot be scaled economically for fusion applications.

2.3 Molten Salt Recovery Process

The molten salt recovery process has shown considerable promise for removing hydrogen isotopes down to the ppm level for liquid lithium [28,29]. In this method, liquid lithium and a molten salt, a mixture of lithium halides, are mixed together. The LiT, being a salt-like material, is preferentially distributed to the salt phase. The salt is then separated from the lithium by taking advantage of the large difference in densities. The tritium in the salt can be recovered by an electrolysis process. The dissociation potential of LiT is about 0.9 V, whereas that of the salt is more than 2 V [28]. The tritium is recovered from the molten salt by sweeping a porous stainless steel electrode with a circulating stream of inert gas, with subsequent recovery of the tritium from the inert gas with a getter. Individual steps of the molten salt recovery process have been demonstrated at laboratory scale. The volumetric distribution coefficient of tritium in salt to that of tritium in lithium has been measured as about four. The mixing and separating of the lithium and salt have also been verified. The electrolysis

steps have also been experimentally verified, with more than 90% tritium recovery from the salt when 0.9 V is applied to the salt. There is no salt decomposition. A similar molten salt recovery process has been demonstrated in fission reprocessing [28].

The molten salt recovery process is somewhat complicated with several delicate steps. The complete separation of salt from lithium, and lithium from salt, is required to reduce potential problems that can be caused by activation, corrosion, and salt solidification in the cold part of the coolant loop. Therefore, two secondary separation units may be required to further separate lithium from salt, and salt from lithium. This will reduce the mutual entrainment, but will not change the amount of material carried over by mutual dissolution.

The operation of the purged electrolysis unit is a key step. The tritium released at the electrode has to be rapidly removed from the salt before it has a chance to combine with the lithium. A certain amount of halide may be carried away by the tritium stream by entrainment or as a result of evaporation. The halides must be completely removed before the tritium stream can be combined with the plasma exhaust tritium stream. However, there is no fundamental reason why these steps cannot be demonstrated in an engineering system. The cost of the tritium recovery system should be within the acceptable range for fusion applications. The major advantage of the molten salt recovery process is that it does not require the addition of hydrogen, as do some other systems. Therefore, a costly step of separating hydrogen isotopes may not be required.

The molten salt recovery process is the only lithium/tritium recovery concept demonstrated experimentally to a tritium concentration level less than 1 wppm. Therefore, basic information is available to define the system. The condition of the tritium stream, such as flow rate, H/T ratio, and impurity concentrations can all be determined.

3. EVALUATION OF TRITIUM RECOVERY METHODS

Four methods of tritium recovery from liquid lithium are described in Section II: yttrium gettering, permeation combined with oxidation, permeation with cold trapping, and molten salt extraction. Presented in this section are a comparative evaluation of the methods and the selection of a reference method.

3.1 Extraction Methods - Evaluation

To assess the advantages and disadvantages of different tritium extraction methods for liquid lithium, the ideal requirements for an extraction system were defined. These requirements included both the condition of the lithium after extraction and the technical and engineering characteristics of the extraction method. Our goal was to have the reference extraction method meet as many of these requirements as possible. The acceptability of the different extraction methods was matched to how well a given method met these requirements.

The criteria used to evaluate each of these tritium extraction systems include scientific feasibility issues, engineering feasibility issues, and economic considerations. The criteria were:

1. scientific basis of the methods;
2. low tritium inventory in the extraction system - < 0.1 kg [concentration < 1 wppm];
3. extraction system forgiving of temperature transients;
4. extraction system forgiving of high nonmetallic impurity levels - oxygen, nitrogen, etc.;
5. extraction system capable of operation when the reactor is nonoperational;
6. low tritium losses from the extraction system;

7. extraction system capable of removing metallic impurities;
8. minimal use of energy for the extraction system;
9. simplicity of operation of the extraction system;
10. maintenance relatively simple for the extraction system; and
11. minimal capital cost of the extraction system.

These criteria are described below.

Scientific Basis - The scientific basis of each method must be sufficiently developed so that a given method is scientifically feasible. For the four extraction methods the level of scientific work varies considerably. Most of the experimental work has examined the tritium extraction capability under ideal conditions.

For yttrium getters, the yttrium can extract tritium from the lithium; however, recovery of tritium from the yttrium has not been demonstrated. In addition, it appears that to attain 1 wppm tritium in liquid lithium, massive amounts of yttrium will be required (see Sec. II.1). The permeation/oxidation method has not been demonstrated in the laboratory. For the permeation/cold trap method, cold trapping from sodium has been demonstrated in a fission breeder reactor [29]; however, the combination of a permeation window and cold trapping from sodium [25] has not been demonstrated. The two steps in the molten salt extraction process, separation into the salt phase and recovery from the salt phase, have both been demonstrated separately. The combined extraction/recovery from the molten salt has not been demonstrated.

Tritium Inventory - The tritium inventory in the liquid lithium is a function of a method's extraction efficiency. However, the tritium inventory in the extraction system is also dependent on a number of other factors, including the amount of lithium that must be processed per unit time, the mode of operation (continuous or batch process), the number of duplicate extraction units necessary to recover the extracted tritium, the time required to recover

the tritium from the extraction system, and tritium solubility in the extraction system.

Because the yttrium getter body operates in a batch mode, there is a tritium inventory in it. If we assume that the beds are on line 12 hours and that it requires 12 hours to regenerate a bed, then the tritium inventory in yttria beds is 8 kg, equivalent half a day of tritium extraction.

There is a residual tritium inventory in the palladium/zirconia window used for the permeation/oxidation method. With this method, there is additional tritium in the tritiated water collection device, a molecular sieve bed or a cryogenic trap, and in the electrolysis unit. Because these components would function on a batch basis, there would be at least two sets of these components in the plant. The overall inventory in the permeation/oxidation system for a 1000-MW(t) reactor is > 0.5 kg.

The permeation/cold trap method would require a palladium window and a sodium loop with a cold trap. The tritium inventory would be that in the window plus that in the cold trap. Because the cold traps would have to be cycled at least on a daily basis, the minimum inventory would be that in the sodium, 0.1 kg in the cold trap, plus that in the palladium window. The total inventory is estimated at 0.5 kg.

The molten salt extraction system method requires a series of contactors, a series of electrolysis units, and a cleanup train for the gas sparge stream from the electrolysis units. For the first two units, which can operate in a continuous mode, the tritium inventory is minimal, < 0.1 kg. The cleanup train may also be able to operate in a continuous mode. Therefore, the total tritium inventory is < 0.1 kg.

Temperature Transients - Temperature transients are expected to be significant for the permeation/oxidation method and the yttrium getter method, because both these systems assume a given lithium temperature at either the window or the yttrium getter surface. For the permeation/cold trap method, the intermediate sodium loop will buffer any temperature transient effect. The molten salt extraction method should not be affected by temperature transients because here, the molten salt acts as a buffer.

Nonmetallic Impurities - The effect of nonmetallic impurities (nitrogen, oxygen, and carbon) is expected to be extremely important for the two

permeation-based methods and for the yttrium getter method. For all three of these methods, the nonmetallic impurities can poison the active surface over long time periods even if a separate cleanup system to remove these impurities is provided. For the molten salt extraction method, the effect of the nonmetallic impurity is nonexistent for the contactor and can be handled in the electrolysis unit by electrochemical means or by filtration.

Reactor Nonoperational - The operation of the extraction system if the reactor is non-operational was assessed. For the two methods based on permeation, the lithium would have to be maintained at fairly high temperatures to ensure that the methods were efficient. For the yttrium getter method, the lithium could be at low temperature. For the molten salt extraction method, a decrease in the lithium temperature could be handled by using a low-melting salt or by providing heating of the 0.3% of the lithium processed.

Tritium Losses - The tritium losses caused by high-temperature components and high tritium partial pressures during tritium recovery from the four methods was assessed. For the yttrium getter, a temperature of 700°C must be used to efficiently extract the tritium from the yttrium getter. For the LiCl-LiBr-LiF molten salt, which has a high melting point, the extracted tritium is at high temperature, so significant losses are possible if an engineering-design solution is not found for the electrolysis unit. For the methods based on a permeation window, losses after the window will be minimal.

Metallc Impurities - All four methods are sensitive to metallic impurities to different degrees. The effectiveness of the permeation windows may be reduced if metallic impurities plate out. One might place a purification unit before the permeation window to minimize these effects. The yttrium getter surface, which is the coldest point, would also have a problem with metals plating out and diffusing into the getter when the getters are recycled at high temperature. The electrodes in the electrolysis unit of the molten salt extraction system could also be affected by metallic impurities, again, a purification unit could be used prior to the electrodes.

Energy Requirements - The energy requirements of an extraction system during its operation were estimated because we did not have any examples of a complete system for any of the four methods. For the permeation/oxidation method, energy requirements would include pumping losses for the lithium and

for the oxygen stream past the permeation window, refrigeration and/or heating needs for the traps for the tritiated water species assumed to form, and electrolysis needs to convert the tritiated water to molecular tritium. For the yttrium getter method, the energy requirements include pumping losses for the lithium pumped past the yttrium getters and heating needs for recycling the yttrium beds twice daily. For the permeation/cold trap method, the energy requirements are primary and include the heating needs for recycling the cold trap twice daily. For the molten salt extraction method, the energy requirements include pumping losses for the lithium to the contactor and the salt in the salt loop, as well as the electrolysis needs to convert the hydride to molecular tritium. We have estimated that pumping losses would be equivalent for the two permeation methods and the molten salt method. The pumping losses for the yttrium getter method could be high if extremely large amounts of getter are required. The main energy requirements for the permeation/oxidation method, the permeation/cold trap method, and the getter method are the recycling of large volumes of material twice daily. The expected electrolysis needs of the molten salt method are expected to range half to one-tenth those values.

Operation - The two permeation methods depend on a passive mechanism, solid state diffusion, to achieve tritium extraction from the lithium. If the temperature of the lithium decreases, diffusion and extraction will be reduced. If the windows are poisoned, diffusion will cease. Replacement of the windows is not a simple or rapid process. The yttrium getter process also depends on solid-state diffusion to extract tritium. Again, if the surface is poisoned, extraction of tritium may cease. Spare beds of yttrium getter would be needed as replacements. These would be fairly simple to replace, but the cost (thin-foil yttrium is \$23,000/m²) would be very high. The components of the molten salt method are not extremely expensive and are easy to replace because they are small; this method appears to be advantageous.

Maintenance - The permeation/oxidation method requires routine maintenance on the gas stream and the components used to collect the extracted tritium and electrolyze it. Maintenance of the permeation window is required if and when it is poisoned. If metallic impurities are collected on these windows, their gamma radioactivity will increase during their lifetimes. Replacement of this component may be extremely difficult. The yttrium getter

method will require routine maintenance on the getter beds. Because they represent the cold spot in the system, their gamma radioactivity contents and expected to increase during their lifetimes. Shielding will be required. The permeation/cold trap method will require routine maintenance of the sodium intermediate loop and the cold trap. Replacement of the permeation window may be extremely difficult. The small components of the molten salt method are designed as easily replaced modular units. Maintenance on any one of these molten salt components should be fairly easy.

Capital Cost - We considered that each method would require a gaseous cleanup system and an isotope separation system; thus, we did not factor these costs into our evaluation. Rather, we addressed the unique components of each method.

The major costs of the permeation/oxidation method are for the permeation window (estimated cost for one Pd/Zr window is \$6 million [27], and the tritiated water collection system and electrolysis system (\$10 million) thus, the total estimated cost is \$16 million. The major costs of the yttrium getter method are for the yttrium getter beds. We estimate that 5200 m² of yttrium, equivalent to the surface area of the permeation window, is needed, for a material cost of \$130 million. The major costs of the permeation/cold trap method are for the permeation window (\$5 million), the intermediate sodium loop (\$10 million), and the cold traps (\$10 million); thus, the total estimated cost is \$25 million. The major costs of the molten salt method are the contactors (\$5million) and the electrolysis units (\$10 million), for a total estimated cost of \$15 million. The permeation/oxidation and molten salt methods have comparable costs.

3.2 Reference-Method Selection

The four methods, permeation/oxidation, yttrium getters, permeation/cold trap, and molten salt were evaluated to determine how well each met the requirements of an ideal tritium extraction system for liquid lithium. A summary of this evaluation is shown in Table 2. We have placed equal weight on each of the requirements. A maximum value of 10 is given if a method achieves the desired requirement. A value of 0 is given if a method does not address a requirement. A value of 5 is given if a method has average performance for a given requirement.

Table 2
An Evaluation of Four Methods to Extract Tritium from Liquid Lithium

Requirement/	Method			
	Yttrium	Perm/Ox	Perm Trap	Molten Salt
1. Scientific feasibility	Yes(10)	Yes(10)	Yes(10)	Yes(10)
2. Inventory (kg)	0.1(10)	<0.5(5)	0.5(0)	<0.1(10)
3. Transient sensitivity	High(0)	High(0)	Low(10)	Low(10)
4. Impurity sensitivity	high(0)	high(0)	High(0)	Low(10)
5. Nonoperational reactor	Yes(10)	No(0)	No(0)	Yes(10)
6. Tritium loss (Ci)	High(5)	Low(10)	Low(10)	High(5)
7. Metallic impurity sensitivity	Yes(5)	Yes(5)	Yes(5)	Yes(5)
8. Energy requirements	High(5)	High(5)	High(5)	Medium(10)
9. Operation	Medium(5)	Medium(5)	Medium(5)	Easy(10)
10. Maintenance	Medium(5)	Hard(0)	Hard(0)	Easy(10)
11. Capital cost	High(0)	Low(10)	Medium(5)	Low(10)
Overall Rating	55	50	50	100

We found that the molten salt extraction method could achieve most of the requirements we considered necessary in an engineered system. Therefore, this method was the one used to evaluate the gas streams that would arise from a tritium extraction method for liquid lithium.

4. REFERENCE SYSTEM

The reference system incorporated a self-cooled liquid lithium blanket with molten salt extraction of tritium. The reference parameters are given in Table 3. The system consists of three fluids: lithium, salt, and sparge gas. The sparge gas, which comes from the electrolyzers, is the tritium stream to be sent to the blanket processing system.

A flow sheet for the three fluid systems is given in Figure 6. The lithium circulates through the blanket and first wall, and the temperature at the outlet of the blanket is 500°C. A portion of the coolant stream then passes through a heated zirconium bed to remove oxygen. A portion of the stream (0.3%) is then passed to the tritium recovery system, the molten salt contactor. The coolant then flows through the intermediate heat exchanger to transmit the thermal power to the intermediate sodium loop. The lithium coming out of the intermediate heat exchanger is at 300°C. A portion of the cold stream is passed to a cold trap that removes certain impurities such as carbides and metals. After passing through the heat exchanger and the cold trap, the lithium from the cold leg is returned to the blanket. The lithium loop will also contain a number of subsystems not shown in Figure 6, including supply tanks and dump tanks, a helium cover gas, filters, and a considerable number of valves. Each one of these components is a potential source of impurities. A significant point is that many of the impurities in the lithium will activate in the neutron environment.

The salt loop includes the contactors, the electrolysis unit, and the salt purification station. The salt passes through the primary contactor where it extracts tritium and impurities from the lithium. Then the salt is passed through a second contactor, followed by the electrolysis unit. Tritium plus impurities are evolved at a sparged electrode, and the product tritium is carried in the sparge gas. After electrolysis, the salt passes through a purifier that consists of a halogen (Br_2) sparging unit and a cold trap. The halogen (Br_2) sparge converts oxide impurity (Li_2O) to halide salt (LiBr). Then the salt flows back to the primary contactor.

The sparge gas is the blanket outlet stream of primary interest in this study. The gas, primarily helium plus the tritium product, flows to the Blanket Processing System. After processing and removal of the tritium, the helium sparge stream flows back to the electrolysis unit. The evaluation involves identifying and quantifying source terms and equilibrium levels for

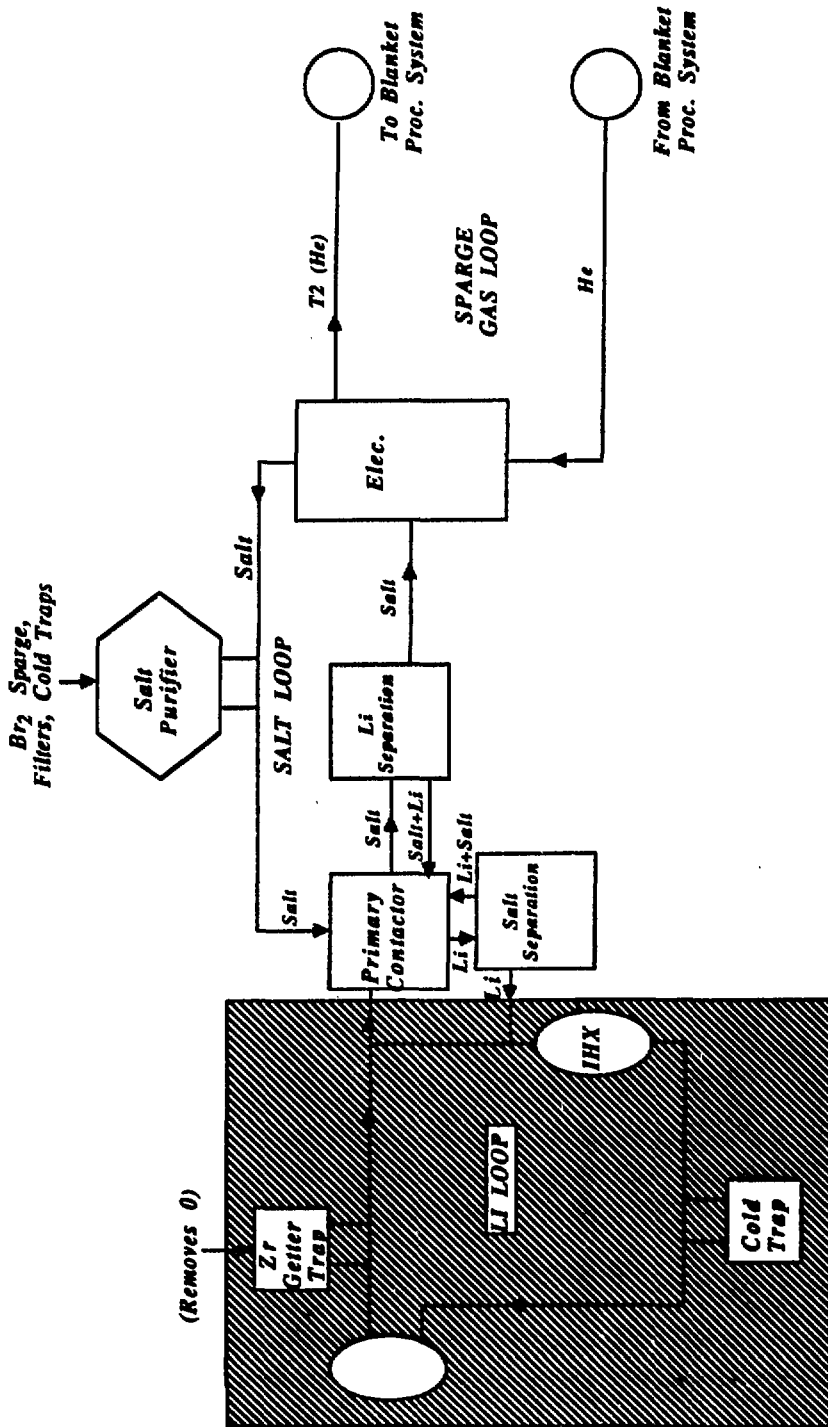


Figure 6 Flow sheet for blanket tritium systems.

Table 3
Reference Parameters for the Blanket Tritium System

Breeding medium	Liquid lithium
Coolant	Liquid lithium
Secondary coolant	Sodium
Structural material	HT-9
Thermal power	1000 MW
Thermal energy/fusion potential	20 MeV
Breeding ratio	1.1
Tritium recovery method	Molten salt extraction
Lithium flow rate (blanket)	1190 kg/s = 4.28×10^6 kg/h
Lithium inlet temperature	300°C
Lithium outlet temperature	500°C
Tritium concentration in Li	1 wppm
Tritium bred per day	150 g
Tritium flow to extractor	12,500 kg/h = 0.3% of coolant flow
Tritium inlet concentration (Li)	1.0 wppm
Tritium outlet concentration (Li)	0.5 wppm
Salt flow to extractor	50,000 kg/hr
Tritium inlet concentration (salt)	0.4 wppm
Tritium outlet concentration (salt)	0.5 wppm
Electrode sparge rate	66,000 L/h at 500°C
Tritium level (T_2) in sparge gas	10^{-3} atm = 100 Pa
Salt composition:	
LiF	22 mole %
LiCl	31 mole %
LiBr	47 mole %
Density of salt	2.1 g/cm ³

impurities in lithium, including radionuclides. The transport rates to the salt and to the sparge gas are then determined. The specific procedures are:

1. Identify and quantify the sources of protium contamination in the lithium: these include the HT-9 structure, residual amounts in the lithium, the cover gas, residues from manufacture, back-diffusion from the steam generator, and inleakage of air.
2. Review the available data from operating experimental liquid lithium loops and sodium-cooled reactors to obtain information on impurity levels.
3. Identify and quantify the sources of other contaminants in the lithium: including nonmetallic elements (e.g., O, N, C, P, S, F, Cl, Br, etc.), metallic elements (Fe, Cr, Na, K, Ni, Si, Ca, Al, etc.), and particulates.
4. Calculate the rates of activation of the lithium and impurities contained therein.
5. Calculate the rates of transport of the various species - impurities, radionuclides, and particulates - from the lithium into the salt.
6. Determine the transport of impurity species from the salt to the sparge gas.
7. Define the composition of the sparge gas stream.

5. EVALUATION OF TRANSPORT RATES OF IMPURITIES

5.1 Source Terms for Protium (H) in Lithium

Hydrogen isotopes are unique in their ability to permeate through metal surfaces. Thus, substantial amounts of protium can enter the lithium from indirect sources, such as from the intermediate sodium loop of the heat transfer system and from the plasma. Other sources of contamination arise from direct contact, such as with the HT-9 structure, residues present in the lithium, the cover gas, air inleakage, the salt, and various residues from manufacture.

5.1.1 Permeation of Protium from the Intermediate Sodium Loop: There are substantial quantities of protium that come into the intermediate sodium loop through permeation and/or leakage through the steam generator. The steam oxidizes the HT-9 cladding in the steam generator, producing hydrogen (protium) gas by a reaction such as



The hydrogen gas is initially produced as atoms, which can readily be absorbed into the HT-9 tube walls and then permeate into the sodium. This mechanism has appeared to be a major source of protium in the sodium of LMFBRs [27-30]. Scaling from UWMK-I [12], the area of the steam generator is 4.54×10^3 , cm², including scaling for power and thermal conductivity of the steel. If the oxidation of the steam generator consumes one mil per year, then the hydrogen produced is 5.4 g/h, assuming the reaction above.

Leakage of water or steam into the lithium or the sodium intermediate coolant must be prevented. Based on criteria from sodium systems [28,31,32], when a leak of greater than 0.1 g/s H₂O is detected, corrective action is taken. For purposes of estimating the magnitude of this effect, the time-averaged leak could be 0.01 g/s. This translates to a source of 4 g/h.

In LMFBR systems, the source from corrosion is generally considered to dominate the H source term [33]; accordingly, the source of protium into the Na is estimated to be 5.4 g/h. The amount of protium that gets into the lithium is controlled by the rate of permeation through the intermediate heat

exchanger. The intermediate sodium loop is assumed to have a cold trap, which can maintain a H_2 over pressure of 0.01 Pa (10^{-4} torr) [25,28]. The intermediate heat exchanger is assumed to have a surface area of 5000 m^2 and a wall thickness of 1.5 mm. The average temperature is 400°C. The permeation rate of protium through the HT-9 into the lithium is calculated to be 9.0 g/d, using a permeability of 5×10^{-3} $\mu\text{mol H} / (\text{s}\cdot\text{m}\cdot\sqrt{\text{kPa}})$ [34].

5.1.2 Injection of Deuterium and Hydrogen from the Plasma through the First Wall: Tritium injection into the first wall coolant has been calculated to be in the range about 10^3 to 10^5 Ci/d. To establish a base case, calculated rates from the International Tokamak Reactor (INTOR), phase II A are used [35]. For that case, tritium injection rates were calculated to be about 0.2 g/d. For 1000 MW, that rate would be 0.3 g/d tritium, or 0.2 g/d (0.1 g-atom/d) deuterium. The protium content of the plasma is estimated to be one or two percent. On the assumption that the protium in the plasma is two percent, the calculated injection rate is 0.008 g-atom H/d.

5.1.3 Production of H from (n,p) Reactions in the Structure: Production of H^2 and He gas were discussed in UWMAK-I [12]. Scaling from calculations in that report (p. VI-C-45), production rate in the first wall is 330 appm/(MW-yr/ m^2) for Fe in HT-9 and 36 appm/(MW-y/ m^2) for Cr in HT-9. Total H production is estimated to be 400 appm/(MW-y/ m^2) for the HT-9. Assuming a neutron wall loading of 5 MW/ m^2 , production per year of H from nuclear reactions in the HT-9 is 2000 appm, or 36 wppm (0.1 wppm/d). Assuming this production rate in the blanket, and that the HT-9 mass is 1×10^5 kg, the daily production rate of H is 10 g.

5.1.4 H Residue in the Lithium: This particular source starts out with the initial H content of the lithium, estimated to be 100 wppm. This amounts to a H inventory of 30 kg. On the assumption that 1/3 of this excess H inventory is removed each day, this source decreases by a factor of ten each week. The amount extracted is 10 kg the first day, 500 g after a week, 50 g on the thirteenth day, and 1 g after three weeks.

5.1.5 H Residue in the Structure: This source term also varies with time, but in a complicated manner. H on metal surfaces is quickly sorbed into the lithium. By contrast, H in the bulk of the metals must diffuse through the bulk of the metal. The term will quickly decrease in value and eventually it will become virtually constant. It is estimated that the initial value is

1100 g H the first day, 100 g the eighth day, 20 g the twentieth day, and a constant value of 10 g per day after 30 days.

5.1.6 Miscellaneous: A number of other sources of hydrogen into the lithium exist. Hydrogen streaming back from the salt is not considered here because it is discussed as a part of the salt evaluation. Air leakage into the cover gas over the lithium is a finite source but the amount of H is expected to be small compared with the sources identified above. Other possible sources are assumed to be negligibly small and are not considered further.

5.1.7 Total Source of H into Li: The total amounts of protium (and deuterium) sources in the lithium are summarized in Table 4. The results of the analysis predict that the initial protium level will be about 75 wppm. This level will fall to about 10 wppm with in a week and will decrease further to about 0.2 wppm after one month.

Table 4
Sources of H into Li

Source	Rate, g/d	H/T atom ratio	wppm H
Permeation from sodium	9.0	0.18	0.06
First wall (D)	(0.2)	(0.002)	
First wall (H)	0.008	--	
(n,p) reactions in HT-9	10	0.2	0.1
Residue in Li	10,000-0	200-0	Time dependent
Leaching from HT-9	1,100-10	22-0.2	Time dependent

Total (steady state)	29	0.6	0.20
Maximum	11,100	222	75

5.2 Other Lithium Contaminants - LMFBR and Lithium-Loop Operation

To obtain information on impurity levels in operating systems, data reported for LMFBRs [36-46] and experimental lithium loops [47-74] was compiled. The results are summarized in Tables 5 and 6. A few observations can be made from these two tables.

1. The larger the liquid metal system is, the lower the impurity levels are. This implies that source terms - leakage, outgassing, dissolution, etc. - scale nonlinearly with size.
2. Impurity levels in sodium are lower than those in lithium. The ratio is less than 7/23, which factor directly puts wppm numbers on the same relative atomic basis. The differences are due to two factors: (1) the greater reactivity of lithium and (2) the relative efficiency of purifying sodium by cold trapping.
3. Major nonmetallic impurities are components of air (oxygen and nitrogen), hydrogen, and carbon.
4. Major metallic impurities are constituents of the structural alloys present.

Table 5
Observed Impurities in Operating LMFBRs, wppm

Reactor Power, MW(t) Location Volume, L Structure	FFTF 400 USA 5×10^5 SS	JOYO 75 Japan 1000 SS	BN350 USSR	PFR UK	CAPHE Loop France	Phoenix 600 France	EBR-II 62.5 USA Croloy
H	0.04	5		5.0	0.05	0.1	0.1
O	0.5	10	5-8	10		< 1	1.1
N		50	2-5				
C	0.06	30	20-30		20		0.2
B	0.1					< 0.05	
Bi						2.9	
P	0.3						
S	0.1						
Cl	0.15	30	7-10				
F	<0.1						
Br							
I							
As							
Se							0.2
Si	0.20						37
Sn							
K	292						
Be							
Mg	0.05						
Ca	0.14						
Sr	< 0.01						
Al	0.05						
V	< 0.01						
Cr	0.14	5					
Mn	0.02						
Fe	2.05	10					0.07
Ni	0.18	5					
U	< 0.001						

Table 6
Observed Impurities in Operating Experimental Lithium Loops, wppm

Facility/ Location	LPTL/ANL USA	ELS/HEDL USA	V/ANL USA	SCK/MOL Belgium	UW USA	ORNL USA	Austria
	Li	Li	Li	Li	Li	Li	Li
Volume, L	200	3800	0.5	10	10	1	1
Structure	304 SS	SS	V-15 Cr	316 SS	316 SS	SS	SS
Max T, °C	500	430	600	500	450	600	600
H	100	50	110				
O	100 ^a	25		~100	275	100	
N	400	20	13 ^b	10 ^b	80	50	100
C	5	2		4		50	
B							
P							
S							
Cl						5	
F							
Br							
I							
As							
Se							
Si		350					10
Na						5	3000
K							5
Be							
Mg							
Ca		100 ^c				10	≥ = 10
V							
Cr	20				7		2
Mn							
Fe	50				7	10	5
Ni	20				< 5		4
U							

^aIncludes solid particulates.

^bGettered with Zr or Ti to remove N.

^cTime dependent (decreasing with time) - value is early average.

5.3 Assessment of Amounts of Other Impurities in Lithium, Including Nonmetallic Elements, Metallic Elements, and Particulates

For species other than hydrogen isotopes, the sources of impurities in lithium are from direct contact, such as the molten salt, the structure, the cover gas, or inleakage of fluids. In the case of protium (H) contamination, it was possible to quantify the various source terms and then calculate the steady-state level. For the other impurity species (nonmetallic elements, metals, and particulates) there is not a sufficient data base to quantify the source rates. Therefore, approach taken is to estimate the probable steady-state concentration levels of nonmetallic elements in the lithium. Transport rates into the salt are then determined based upon these concentration levels. The sources of contamination include the molten salt, the HT-9 structure, the cover gas, and residual contaminants on surfaces. Considered below are the sources of nonmetallic elements, metallic elements, and particulates in the lithium.

5.3.1 Nonmetallic Elements in Lithium: Important sources of nonmetallic element impurities include the halide salt, the HT-9 structure, organics, air, and water. Each of these sources is assessed below.

5.3.1.1 Contamination of the lithium by the molten salt. The major concern is the dissolution and the entrainment of the halide salts into the lithium. The salt is expected to reach saturation solubility in the lithium. The saturation solubility is less than 100 appm for the lithium halides [28]. Assuming that the entrained salt can be reduced to low levels, the amount of lithium halides is assumed to be 100 appm total, or 22 appm LiF (60 wppm F), 31 appm LiCl (160 wppm Cl), and 47 appm LiBr (540 wppm Br). In general, nonmetallic elements are preferentially extracted from the lithium into the salt. In addition, as discussed in further detail later, the nonmetallic elements are removed from the salt by electrolysis [22] or by halogen sparging [75]. Therefore, source terms of nonmetallic elements other than halides from the salt need not be considered further. Source terms into the salt are considered separately. It is useful to note that most metallic impurities, cations in the salt, will be reduced by the lithium metal and preferentially extracted into the liquid lithium phase.

5.3.1.2 Leaching of nonmetallic impurities from the HT-9 into the lithium. The chemical composition of HT-9 [76] is given in Table 7. Major

nonmetallic components are C, P, and S. These nonmetallic elements will tend to be slowly extracted into the liquid lithium. The C is present as an integral part of the alloy, forming stable ferrites. The form of C in liquid lithium is as the acetylide, nominally C_2^{2-} . Owing to comparatively high stability in the steel and the comparatively low stability in solution, the amount of C that dissolves into the Li is comparatively small. By contrast, the S and P are present as impurities in the steel and form simple salts in solution. Therefore, the fractions of P and S that dissolve are comparatively high. It is estimated that the steady-state levels of C, P, and S are 5, 10, and 5 wppm in the lithium, respectively, on the assumption that the HT-9 is the only source of these elements.

Table 7
Chemical Composition of HT-9 (Sandvik)^a

Element	Composition, wppm	
	In HT-9	In Lithium
C	2000	5
Si	4000	10
P	200	10
S	100	5
Mn	5500	
Cr	1.15×10^5	
Ni	5000	
Mo	1.0×10^4	
W	5000	
V	3000	
Fe	Balance	

^aHandbook of Fusion Reactor Materials [77]

5.3.1.3 Miscellaneous sources of nonmetallic elements in lithium. During operation, there is a potential for inleakage of air into the lithium or the cover gas above the lithium. In principle, there may be a finite source of organic lubricant; although, design goals are to prevent any

contact of organics with lithium. Similarly, the current designs aim to ensure that lithium cannot contact water or air in any form. At present, there is no data base on operating with large volumes of liquid lithium under prototypical fusion conditions. Also, in addition to direct leakage, the metal surfaces prior to adding lithium will have some residues. The impurities considered herein are air components, organics, and water. The principal nonmetallic elements from these sources are H, N, O, and C. To estimate the magnitude of these sources, experience from operating sodium-cooled LMFBs and lithium loops is used (see Tables 5 and 6 above).

5.3.1.4 Summary: Nonmetallic elements in lithium. The typical values for nonmetallic elements H, O, N, and C are tabulated for existing lithium and sodium systems in Table 8. Because of the mass difference, the wppm values for sodium are multiplied by a factor of 23/7 to put the data on a comparable atomic basis with lithium. The values for sodium are comparatively low as a result of two factors: (1) the relatively lower stability of sodium compounds and (2) the effectiveness of cold trapping as a method of purifying sodium on line. In lithium, the cold traps and the getter traps can lower O, N, and C only to about 100 wppm. However, these nonmetallic elements are preferentially extracted into the salt, and the nitrogen and carbon are removed from the salt by electrolysis. There is evidence that the molten salt extraction process may be particularly effective for removing nitrogen from lithium [73]. The oxygen levels in the salt can be controlled by halogen sparging [75] or possibly by cold trapping. It is assumed that salt purification will keep the oxygen level in the salt at a level not higher than 50 wppm. This level translates to 25 wppm in the lithium. The protium level, calculated above, is comparatively low, owing to efficient removal from the sodium and by the molten salt extraction. The values for nitrogen and carbon in lithium are assumed to be at the lower end of the range of what is achievable in current systems.

5.3.2 Metallic Impurities in Lithium: Using the same procedures as above, the levels of metallic impurities in the lithium were estimated, using data from existing liquid metal systems as a basis. The results are summarized in Table 9.

Table 8
Typical Impurity Levels of Nonmetallic Elements in Liquid Metal Systems

Element	CONCENTRATION, wppm			
	Lithium Loops	LMFBR in Sodium	Equivalent from LMFBR in Lithium	Estimated Level in Lithium
H	50	0.05	0.2	0.2
O	25-100	0.5	1.6	25
N	> 20	1 ?	3 ?	10
C	4	0.05	0.2	4

Table 9
Concentration of Dissolved Metallic Elements in Lithium

Element	HT-9 Structure	SS-Lithium	LMFBR (equivalent)	In Lithium est.
Fe	8.8×10^5	5-20	0.5-1.0	10
Si	4000	10-350	0.6	10
Mn	5500		0.06	1
Cr	1.15×10^5	2-20	6.2	10
Ni	5000	4-20	0.6	5
Mo	1.0×10^4			0.1
W	5000			0.001
V	3000		< 0.03	0.1
Na		5		5
K		5		5
Be				0.1
Mg			0.15	1
Ca		10-20	0.45	5
Sr			< 0.03	0.1
Al		10	0.15	1
U			< 0.003	< 0.001

5.3.3 Particulates in Lithium: There will be some particulates in the lithium. These arise from erosion of the structure and are comprised of metals and metal compounds such as oxides. These species in the reactor may be highly radioactive. The amounts of these materials are rather uncertain. The cold trap on the cold leg of the lithium loop has a wire mesh that will filter out some of this material. It is estimated, on the basis of Lithium Processing Test Loop (LPTL) experience at ANL [29,67,68,72,73,74], that this material has an equivalent concentration the order of 100 wppm and that the material is primarily metal-oxygen composite.

5.4 Activity Levels in Lithium

The above impurity levels in lithium were used to calculate activation levels for these impurity species. The basis of the calculation was a 100-group neutron spectrum from UWMK-I [12] at a location 25 cm into the blanket, multiplied by a factor of 3.6 to make the calculation for a neutron wall loading of 3.6 MW/m^2 . The irradiation time was taken to be one year and the activities in curies and the hard gamma dose in rads were calculated, assuming 24 hours decay. The results of the calculation are given in Table 10. Dose rates for beta emitters (C-14, Mg-37, etc.) were not calculated, because the system is assumed to be contained.

Table 10 Activity Levels in Lithium

Reaction	LEVEL				
	wppm in Li	mCi/mg	mCi/kg Li	mR/mg	mR/kg Li
N-14(n,p)C-14	10	1.88E-3	0.019	0.00	0.00
Na-23(n,g)Na-24	5	0.82	4.1	695.	3475.
Mg-24(n,p)Na-24	1	2.86	2.9	2421.	2421.
Al-27(n,p)Mg-24	1	000	0.0	00000	0.00
Al-27(n,a)Na-24	1	1.975	2.0	1673.	1673.
Al-27(n,2n)Al-26g	1	1.7E-7	0.0	6.5E-5	0.0
P-31(n,p)Si-31	10	0.0153	0.15	0.0000	0.0
(est)P-31(n,g)P-32	10	0.035	0.35	0.000	0.0
(est)S-32(n,p)P-32	5	0.17	3.5	0.000	0.0
Cl-35(n,p)S-35	160	4.90	784.	0.00	0.0
Cr-50(n,g)Cr-51	10	0.40	4.	2.56	25.6
Mn-55(n,g)Mn-56	1	0.041	0.041	15.0	15.0
Mn-55(n,2n)Mn-54	1	5.43	5.43	931.	931.
Fe-54(n,p)Mn-54	10	0.52	5.2	88.5	885.
Fe-54(n,a)Cr-51	10	0.11	1.1	0.70	7.0
Fe-54(n,2n)Fe-53	10	000	0.0	000	0.0
Fe-56(n,p)Mn-56	10	0.004	0.04	1.3	13.0
Fe-58(n,g)Fe-59	10	0.01	0.1	2.42	24.2
Ni-58(n,p)Co-58	5	12.25	61.25	2076.	10380.
Ni-58(n,2n)Ni-57	5	0.082	0.41	26.6	133.
Ni-60(n,p)Co-60	5	0.116	0.58	60.	300.
Ti-46(n,p)Sc-46	1	0.69	0.69	285.	285.
Ti-47(n,p)Sc-47	1	0.32	0.32	7.7	7.7
Ti-48(n,p)Sc-48	1	0.76	0.76	520.	520.
Co-59(n,g)Co-60	0	4.2	0.0	2158.	0.0
Co-59(n,p)Fe-59	0	1.3	0.0	327.	0.0
Cu-63(n,g)Cu-64	0.01	4.37	0.04	174.3	1.7
Cu-63(n,a)Co-60	0.01	0.08	0.0	40.5	0.4
Au-197(n,g)Au-198	0.0001	44.9	0.0	3715.	0.4
W-186(n,g)W-187	0.001	2.86	0.0	297.	0.3
U-238(n,f)	< 0.001	9.4	0.0	1402	< 1.4
U-238(n,g)U-239	< 0.001	18.1	0.0	812.	< 1.
U-238(n,2n)U-237	< 0.001	6.94	0.0	141.1	< 0.1

6. TRANSPORT OF IMPURITIES IN THE MOLTEN SALT CYCLE

In Section 4, Figure 6 illustrates the flow paths of the salt. The parameters for the salt system are given in Table 3. As shown in Figure 6, the lithium and the salt are mixed in the main contactor and then separated into the two phases. Because some of the salt is entrained in the lithium, a second-stage separator is used to reduce this cross contamination before the lithium is returned to the lithium loop. The salt also passes through a second-stage separator to remove residual amounts of lithium droplets. The salt is then passed to an electrolysis unit, where lithium is plated out on the cathode and the tritide ion is oxidized to tritium gas, which is collected from the sparged anode. Along with the tritium, H_2 gas as well as NH_3 and C_2H_2 are released to the helium sparge gas. This sparge gas stream is the blanket stream that is the focus of our attention -- it is discussed further later in this report. After passing through the electrolysis unit, the salt stream is purified to remove oxygen and other impurities, including particulates. The purification of the salt needs to be studied in detail, but it is likely to involve (1) halogen sparging to convert oxides to halides [75], (2) cold trapping to precipitate out a variety of dissolved species, and (3) filtering and/or skimming to separate out debris. After purification, the salt is sent back to the lithium contactor to begin the cycle again. The major source of impurities, particularly nonmetallic elements, is the lithium. Other sources (residues in the salt, leakage, and dissolution from the salt containment) are considered to be small in comparison with the lithium source.

6.1 Transfer of Impurities from the Lithium to the Salt

During contacting of the two fluids, tritium and other hydrogen isotopes are preferentially extracted into the salt phase. Volumetric distribution coefficients (ratios of concentrations in salt to those in lithium) for hydrogen isotopes have been measured and are about 3 to 4, owing to the salt-like nature of the hydride [28]. Volumetric distribution coefficients are as high as 10 for nitrogen and about 2 for carbon [29]. Volumetric distribution coefficients for other nonmetallic elements are expected to be at least 2, owing to the salt-like chemistry of lithium compounds with nonmetals. The volumetric distribution coefficient for metals is likely to be substantially

less than 1 for zero-valent species. Further, cations present in the salt will have a very strong tendency to be reduced to the zero-valent form by the lithium metal. Thus, the distribution for metals dissolved in either the salt or the metal lithium will favor the metal lithium. It is estimated that the volumetric distribution coefficient for dissolved metals is about 0.4. Particulates suspended in the lithium will tend to distribute into the salt. Because the particulates in general will have a density much greater than lithium, these particulates will tend to favor the salt phase. It is estimated that the volumetric distribution coefficient for these species is about 2.

The volumetric distribution coefficients are then used to calculate the concentrations of impurities in the salt. The results are shown in Table 11. Of particular interest are the nonmetallic species, those which could evolve gaseous species at the electrodes.

6.2 Radionuclides in the Salt

Sources of radionuclides and quantities of each are given in Table 10. The amounts of each of these species transported to the salt were calculated by the same methods as in Section VI.1. The radionuclides in lithium, the distribution coefficients, and the calculated activities in the salt are given in Table 12. The major sources of activity are gamma-emitters that are metals, e.g., Co-58. Certain nonmetallic radionuclides are present, notably S-35, P-32, and C-14. These are all beta emitters, the radiation does not penetrate, and therefore the dose is zero for these species. There is some possibility of evolution of radionuclides during electrolysis, but the amounts are expected to be small, and the possible species are only beta emitters. There is no evidence that gamma emitters will be evolved.

6.3 Electrolysis

The tritium, in the form of T_2 gas, is released from the sparged electrode. The electrolysis involves oxidation of the tritide ion to zero-valent tritium gas. The electrolyzed salt has been mixed with lithium and it has unit activity of lithium, i.e., it is strongly reducing. Any tendency to form H^+ is strongly suppressed by the reducing conditions of the salt. This means that hydrohalic acids (HCl, HF, HBr) are not chemically stable in the

Table 11
Transfer of Impurities from the Lithium into the Salt

Element	Estimated Level in Li, wppm	Volumetric Dist. Coefficient		Conc. in salt wppm	Species Elec.	x/T Atom Ratio
		Meas.	Used			
H	0.10(calculated)	3	2	0.1	H ₂	0.6
D	0.002	4	2	0.0005	HD	0.003
T	1.00	4	2	0.5	HT	1.000
O	25		2	11.5	--	5.
N	10	10	2	4.6	N ₂	2.2
C	5	2	1	1.2	C ₂ H ₂	0.6
P	10		2	5	PH ₃	1.0?
S	5		1	1.2	--	0.2
F	60		-	-		
Cl	160		-	-		
Br	540		-	-		
Si	10		1	2.5	SiH _x ?	0.5
Mn	1		0.4	0.1		
Cr	10		0.4	1.0		
Ni	5		0.4	0.5		
Mo	0.1		0.4	0.01		
W	0.001		0.4	0.0001		
V	0.1		0.4	0.01		
Na	5		0.4	0.5		
K	5		0.4	0.5		
Be	0.1		0.4	0.01		
Mg	1		0.4	0.1		
Ca	5		0.4	0.5		
Sr	0.1		0.4	0.01		
Al	1		0.4	0.1		
U	< 0.001		0.4	< 0.0001		
Crud	100		2	50		

Table 12
Radionuclides in the Salt

Species	T _{1/2}	mCi/kg Li	mR/kg Li	Dist. Coeff.	mCi/kg Salt	mR/kg Salt	Species Elect.
C-14	β ⁻ .53704	0.019	0.00	1	.005	0.00	C ₂ H ₂
Na-24	B ⁻ , 15 h	9.0	7569.	0.4	0.86	721.	-
Si-31	β ⁻ , 2.6 h	0.15	0.0	1	0.04	0.00	-
P-32	β ⁻ , 14.3 d	3.85	0.0	2.	1.83	0.00	PH ₃
S-35	β ⁻ , 87.2 d	784.	0.0	2.	373.	0.00	-
Sc-46	β ⁻ , γ, 43.8 d	0.69	285.	0.4	0.066	27.1	
Sc-47	β ⁻ , γ, 3.3 d	0.32	7.7	0.4	0.030	0.73	
Sc-48	β ⁻ , γ, 43.7 h	0.76	520.	0.4	0.07	49.5	
Cr-51	γ, 27.7 d	5.1	32.6	0.4	0.49	3.1	
Mn-54	γ, 312 d	5.2	885.	0.4	0.50	84.3	
Mn-56	γ, 21.6 h	0.081	28.0	0.4	0.008	2.67	
Ni-57	β ⁺ , 36 h	0.41	133.	0.4	0.04	12.7	
Co-58	β ⁺ , γ, 71 d	61.25	10380.	0.4	5.83	989.	
Fe-59	β ⁻ , γ, 44.5 d	0.1	24.2	0.4	0.01	2.30	
Co-60	β ⁻ , γ, 5.27 y	0.58	300.	0.4	0.055	28.6	
Cu-64	β ⁻ , γ, 12.7 h	0.04	1.7	0.4	0.004	0.16	
Au-198	β ⁻ , γ, 2.7 d	0.0	0.4	0.4	0.00	0.04	
W-187	β ⁻ , γ, 23.9 h	0.0	0.3	0.4	0.00	0.03	

medium. The electrolysis occurs at a potential of about 0.7 V [18] and no decomposition of the halide constituents (which require >2 V) has been observed [28]. There is some tendency of the T₂ to go back into the salt phase, owing to the reducing conditions of the salt. Therefore, it is assumed that only a portion of the tritide (20%) is electrolyzed in one stage. Other species that will be evolved during electrolysis include other hydrogen isotopes, nitrogen, acetylene, and possibly other species such as phosphine, ammonia, silanes, and hydrocarbons. The sparge gas may also contain traces of inert gases that have been dissolved in the salt.

7. CONDITION OF THE SPARGE GAS STREAM

The sparge gas coming out of the electrolysis unit is the blanket tritium product stream that is to be sent to the blanket processing system (the blanket interface). The characteristics of this stream are summarized in Table 13. The major features of this stream are summarized below:

1. The stream is helium gas, plus the tritium product, plus impurities. The concentration of hydrogen isotopes in the stream is about 1000 vppm, or 100 Pa. These products must be extracted from the tritium under conditions where the helium is quantitatively separated from the product. The blanket stream is 99.9% helium, compared to the plasma exhaust stream, which is about 5% helium.
2. The ratio of H to T in the product stream is about one. The implication of this is that the blanket product stream may not require its own isotopic separation system, for a liquid lithium blanket. This is contrasted to other blanket concepts where H/T ratios may be as high as 10^3 to 10^5 .
3. The amount of deuterium in the product stream is very small, about 0.3% of the tritium level.
4. A significant level of nitrogen gas may be present in the sparge stream, levels were calculated to of about 2000 ppm. This gas will have to be separated from the product stream.
5. Owing to the vapor pressure of halides at 500°C and to entrainment, there will be lithium halides present at levels of about 1 vppm in the sparge gas. These species must be removed to prevent corrosion of other processing components.
6. The product stream is expected to contain small amounts of beta emitters, including C-14 and possibly P-32 and S-35.

JANAF data were used to calculate equilibrium amounts of NH_3 and the vapor pressures of the lithium halides [77]. In addition to these species, a significant amount of acetylene could be present. The amount is uncertain because of a lack of data on electrolysis behavior. The acetylene will contain about 0.06 millicuries of C-14 per milligram of C_2HT , as a result of nitrogen activation. This compares to a tritium activity of 1.0 curies per milligram of C_2HT . There are likely to be trace levels of a number of radionuclides in the purge gas, but the analysis to date has not identified a pathway for any gamma-emitters to be present at significant levels.

Table 13
Conditions of Sparge Gas

Sparge gas	He
Sparge gas flow rate	66,000 L/h
Temperature	500°C
Pressure	0.1 MPa(1 atm)
T_2 level, vppm	816.
HT level, vppm	490.
H_2 level, vppm	294.
$\text{D}_2(\text{HD}+\text{DT})$ level, vppm	3.
N_2 level, vppm	2200.
NH_3 level, vppm	0.020
LiBr level, vppm	2.37
LiCl level, vppm	0.03
LiF level, vppm	1.37
$\text{C}_2(\text{HT})_2$, vppm	< 600.?
PH_3	?
S_iH_x	?

8. CONCLUSIONS

The conditions of the gaseous tritium stream from the tritium recovery system of a self-cooled, lithium blanket has been evaluated. The total throughput (dominated by helium), the hydrogen-to-tritium ratio, the corrosion-material composition (mainly halides), and the activation products are all defined from the best available information. The main issues that must be addressed in a mockup blanket exhaust processing system are the following:

1. The total gas throughput in the blanket system is much larger than that of the TSTA (1000 moles/h vs. 15 moles/h). The blanket throughput is dominated by the 10^3 excess of helium sparge gas. Separation of the hydrogen isotopes from helium is feasible. However, the system to be used needs to be defined.
2. The ratio of protium to tritium in the blanket stream is about one. This ratio may be much higher for a very short time at startup, owing to residues of H in the starting Li and HT-9. A study to show the trade-off between the use of a common cryogenic distillation unit with the plasma exhaust and the use of a separate unit to avoid mixing H, D, T needs to be done.
3. The composition and amount of corrosive chemicals in the blanket stream is dominated by the halides. A method to remove the halides to an acceptable concentration is needed.
4. The major radionuclides found were C-14 from nitrogen and P-32 from chlorine. A method to remove the radionuclides to an acceptable concentration, to be defined, is needed.
5. The nitrogen level expected in the blanket stream is much higher than that anticipated for the TSTA plasma exhaust system. A method of removing nitrogen to an acceptable level is needed.

The blanket processing system mockup for TSTA for a self-cooled lithium blanket will incorporate units that can address these issues. The exact configuration of this processing system will be defined in the next stage of this program, as will the interface with TSTA.

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