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TITLE: MIXING RULES FOR AND EFFECTS OF OTHER HYDROGEN ISOTOPES AND OF ISOTOPIC SWAMPING ON TRITIUM RECOVERY AND LOSS TO BIOSPHERE FROM FUSION REACTORS

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MASTER

SUBMITTED TO: 3rd ANS Topical Meeting on the Technology of Controlled Nuclear Fusion (Proceedings) (May 1978)

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A. A. LA-UR-73-1673
MIXING RULES FOR AND EFFECTS OF OTHER HYDROGEN ISOTOPES AND OF ISOTOPIC SHAPING ON TRITIUM RECOVERY AND LOSS TO BIOSPHERE FROM FUSION REACTORS

J. H. Pendergrass

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ABSTRACT

Efficient recovery of bred and unburnt tritium from fusion reactors, and control of its migration within reactors and of its escape into the biosphere are essential for self-sufficient fuel cycles and for public, plant personnel, and environmental protection. Tritium in fusion reactors will be mixed with heavy deuterium and protium via thermal, high-temperature reactions and diffusion into contact zones from steam cycles. Rational design for tritium recovery and escape prevention must acknowledge this fact. Consequences of isotopic mixture are explored, mixing rules for mixed-vent fusion reactor deuterium-lithium lithium are developed, and results are used to extend existing tritium-out effects on reactor recovery and loss models.

INTRODUCTION

Present indications are that at least the first generation of fusion reactors will be fueled with tritium from a heavy isotope enrichment mix. Protections and the large, high-temperature, high-pressure reactor core, will be in the form of a mixture of deuterium, tritium, and lithium. Tritium recovery and tritium loss mechanisms, though similar to those in nuclear reactors, are greatly complicated by the fact that tritium is the second element in the periodic table. Tritium is not only lighter than any other element in the periodic table, but also of relatively low atomic number. Hence, it has the lowest ionization energy and a special affinity for chemical bonds, with the result that it is relatively easy to separate tritium from lithium, and lithium from tritium, and hydrogen from tritium.

Efficient tritium recovery from breeder, burner, and other reactors will be possible only if tritium can be extracted from the reactor core and from the contact zones of the deuterium and tritium mixture, and if the tritium can be transported, concentrated, and recovered with minimum loss.

Work done under the U.S. Department of Energy...
Efficient recovery of bred and unburnt tritium is essential for maintaining low tritium inventories. Escape of tritium to the biosphere can be minimized by maintaining low tritium inventories and by providing engineered barriers to tritium escape. Under emergency conditions, the breeding blanket and the main coolant loops are usually considered to represent the greatest hazard, whereas during normal operation, thin, hot reactor components lying along direct paths to the biosphere are critical to tritium escape. Tritium escapes into these coolant loops and seeps into the biosphere, in general, regardless of the most important function it has for reactor safety. When tritium is released into the environment, it influences the design of recovery systems and the design of the reactor.
Our discussion will be confined to the dilute-solution conditions anticipated for tritium in fusion reactor breeding blankets, coolant loops, structural materials, and perhaps cavity exhaust streams. Early estimates of rates of tritium escape into steam cycles from coolant loops suggested that tritium concentrations in fusion reactor systems might have to be maintained at 1 ppm or less. As the difficulties of tritium recovery from solutions at such concentrations have become apparent, more effective concepts for prevention of tritium escape have been developed, and the importance of potential tritium hazards relative to other potential fusion reactor hazards has been more accurately assessed, so that concentration has been given to operation with tritium concentrations in order of or two higher in magnitude. Nonetheless, dilute-solution theory will still be applicable, or it can be made to apply even where concentrations are still low.

We will focus on the development of mixing rules for equilibrium of high-exposure systems, and on the effects that the presence of a small number of high-exposure systems has on the concentration of tritium in coolant loops, PIPING, BUILDING, and plant-wide operation of tritium recovery systems.

Most of the discussion of fusion reactors will probably be about the water that is used to dilute the tritium in the coolant loops, the water that is used in the tritium recovery system, and the water that is used in the plant. We will also discuss the safety of the other systems. The discussion is all about the amount in the reactor, the tritium inventory, and the processes of the other systems. We will also discuss the implications of the safety of the hydrogen systems.

Many theories have been presented by workers and people. We have seen the work of various people and people. We have seen the work of various people. We have seen the work of various people. We have seen the work of various people. We have seen the work of various people. We have seen the work of various people. We have seen the work of various people.
and then Stewart's law for the solubility of hydrogen isotopes in metals is obtained:

\[ K(H_\alpha) = K(H) \left( \frac{\rho_{H_\alpha}}{\rho_H} \right)^{1/2} \]

Then:

\[ K(H_\alpha) = K(H) \left( \frac{\rho_{H_\alpha}/MT}{\rho_H/MT} \right)^{1/2} \]

where:

\[ K = \frac{\rho}{RT} \]

The use of the empirical constant provides a method for determining the solubility of hydrogen isotopes in metals. The experimental data is binarized rather than with other solutes. Therefore, the following properties other than the solubility of hydrogen isotopes in metals are measured by the process of the metal, and in the absence of the solute. This interpretation should also be noted if the solute is a mixture of hydrogen isotopes, the relative ratio of the solutes in the solution is the constant, e.g., if the solute is a binary, dilute or otherwise.

Thus we conclude that Stewart's law expressions obtained by fitting experimental solubility data for hydrogen isotopes in metals are applicable to individual metal solubilities when hydrogen isotopes are involved. Therefore, for a system containing all three hydrogen isotopes, we write:

\[ K(H_\alpha) = K(H) \left( \frac{\rho_{H_\alpha}/MT}{\rho_H/MT} \right)^{1/2} \]

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In an isotopic mixture there will be isotopic exchange in the gas phase, i.e., in addition to the molecular species \( \text{H}_2/\text{g} \), \( \text{D}_2/\text{g} \), and \( \text{T}_2/\text{g} \), the species \( \text{HD}/\text{g} \), \( \text{DT}/\text{g} \), and \( \text{TH}/\text{g} \) will be present. In addition, the diatomic molecules can dissociate to yield the atomic species \( \text{H}/\text{g} \), \( \text{D}/\text{g} \), and \( \text{TD}/\text{g} \). The following set of six exo-phase reactions are independent:

\[
\begin{align*}
\text{H}_2/\text{g} & \rightarrow \text{H} + \text{H} \\
\text{D}_2/\text{g} & \rightarrow \text{D} + \text{D} \\
\text{T}_2/\text{g} & \rightarrow \text{D} + \text{T}
\end{align*}
\]

\[
\begin{align*}
\text{TD}/\text{g} & \rightarrow \text{D} + \text{T}
\end{align*}
\]

The corresponding equilibria can be expressed in the form:

\[
\frac{2}{P_{\text{H}(g)}} = \frac{2}{P_{\text{H}_2/\text{g}}} \quad \frac{2}{P_{\text{D}(g)}} = \frac{2}{P_{\text{D}_2/\text{g}}} \\
\frac{2}{P_{\text{TD}(g)}} = \frac{2}{P_{\text{TD}_2/\text{g}}}
\]

with similar expressions for the other reactions. Experimental values for the equilibrium constants are listed in Table I.

In terms of \( \gamma_H \), \( \gamma_D \), and \( \gamma_T \), the equilibrium relationships for the gas phase can be written, in the absence of significant molecular dissociation to form the atomic species, in the form:

\[
\left[ \frac{\gamma_H - \gamma_H}{\gamma_H} \right]^2 \left[ \frac{\gamma_D - \gamma_T}{\gamma_T} \right]^2 \left[ \frac{\gamma_T - \gamma_H}{\gamma_H} \right]^2 = K_{\text{TD}}(T)
\]

upon substitution of expression for solution as:

\[
\gamma_{\text{TD}} = \left[ \frac{\gamma_H - \gamma_H}{\gamma_H} \right]^2 \left[ \frac{\gamma_D - \gamma_T}{\gamma_T} \right]^2 \left[ \frac{\gamma_T - \gamma_H}{\gamma_H} \right]^2
\]

Solution for the ratio \( \frac{\gamma_H}{\gamma_H} = \frac{\gamma_D}{\gamma_D} = \frac{\gamma_T}{\gamma_T} = \frac{\gamma_{\text{TD}}}{\gamma_{\text{TD}}} \) is not possible in general. Substitution of the appropriate values for the diatomic species into the solution gives an accurate solution for \( \gamma_H \), \( \gamma_D \), and \( \gamma_T \).

Table I gives an example of the calculation of \( \gamma_H \) from the above expression.

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<th>( T ) (K)</th>
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<th>( \gamma_{\text{TD}}/\gamma_{\text{TD}} )</th>
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*aMoore, et al. (37)*

*bJones (38)*
expressions for \( x_{10} \), \( x_{20} \), and \( x_{30} \) for substitution into the first three equilibrium relations are now of the form:

\[
\begin{align*}
    x_{10} &= \left( x_H \right) \left( x_{10} \right) \left( x_{20} \right) \left( x_{30} \right) \\
    &= \sqrt{\frac{K_{10}^1}{P_{init}^1}} \cdot \sqrt{\frac{K_{10}^2}{P_{init}^2}} \cdot \sqrt{\frac{K_{10}^3}{P_{init}^3}} \\
    &= \left[ \sqrt{\frac{K_{10}^1}{P_{init}^1}} \cdot \sqrt{\frac{K_{10}^2}{P_{init}^2}} \cdot \sqrt{\frac{K_{10}^3}{P_{init}^3}} \right]
\end{align*}
\]

To obtain a numerical comparison with the intuitive mixing rule of Hockin, we consider a mixture of potassium and deuterium of temperature sufficiently low that dissociation into atomic species can be neglected. In this case, it is found that the required average of the trapping coefficients considerably exceeds the maximum potential for tritium.

Based on these approximations, solution for the molecular weight fraction gives:

\[
\begin{align*}
    x_{10} &= \left( x_H \right) \left( x_{10} \right) \left( x_{20} \right) \left( x_{30} \right) \\
    &= \sqrt{\frac{K_{10}^1}{P_{init}^1}} \cdot \sqrt{\frac{K_{10}^2}{P_{init}^2}} \cdot \sqrt{\frac{K_{10}^3}{P_{init}^3}} \\
    &= \left[ \sqrt{\frac{K_{10}^1}{P_{init}^1}} \cdot \sqrt{\frac{K_{10}^2}{P_{init}^2}} \cdot \sqrt{\frac{K_{10}^3}{P_{init}^3}} \right]
\end{align*}
\]

The effect of tritium on both the Wolf and deuterium trapping coefficients is negligible, which leads to the conclusion that the total trapping coefficient is only slightly smaller than that without the trapping rule described above.

The conclusion of this analysis is that, in the presence of tritium, the trapping coefficients for both potassium and deuterium are reduced, but the overall trapping coefficient remains high enough to be beneficial.

The required capacities of tritium recovery and isotopic separation systems are not increased inordinately as a result of isotopic swamping.

Isotopic swamping significantly decreases residual tritium concentrations or enhances the effectiveness of barriers to tritium escape by mechanisms not otherwise available.

The use of tritium recovery methods which are inherently less expensive, e.g., whose capital and/or operating costs are less, is made feasible through isotopic swamping.

These characteristics represent the basis upon which a rule of thumb for a priori assessments of potential beneficial effects of isotopic swamping on fusion reactor tritium recovery methods can be stated. Many processes have been proposed for recovery of tritium from tritium-bearing blanket and coolant-loop steam and unheated tritium from cavity exhaust streams.

The effects of isotopic swamping on all these proposed processes cannot be discounted, and we are forced to concentrate on only a few. In particular, we will discuss effects of isotopic swamping on recovery of tritium by means of tritium window and cold trapping of liquid-liquid trapping reflux and recovering coolant loops and an escape of tritium into the biosphere by permeation through metallic barriers.

Hickman4 examined the effects of isotopic swamping on tritium-window processes for tritium extraction from liquid lithium, which possessed the following characteristics:

- Slow rate of solubility of hydrogen isotopes in metals and Hickman's mixing rule for dissolution of hydrogen isotopes in metals are both applicable.
- The only significant resistance to permeation through the window is the resistance to diffusion of the bulk part of the window membrane.
- Tritium extraction is performed under steady state, isothermal conditions.
Equilibrium with respect to isotopic interchange between diatomic molecular hydrogenic species in the gas phase is attained at all times, the high-temperature limiting values of the isotopic interchange equilibrium constants are applicable, and molecular dissociation to give the atomic species can be neglected. The stated assumptions imply that expressions for the permeation fluxes of protium and tritium through the tritium window can be written in the forms:

\[ J_T = \left( \frac{1}{2} y_T^{\text{total}} - \frac{1}{2} y_T^{\text{total}} \right) \left( y_T^{\text{total}} \right)^{1/2} \]

\[ J_T = \left( \frac{1}{2} y_T^{\text{total}} - \frac{1}{2} y_T^{\text{total}} \right) \left( y_T^{\text{total}} \right)^{1/2} \]

If all these isotope partial pressures are present and if dissociation of the atomic species to give the atomic species in the gas phase cannot be neglected, then the analysis will be more complex, but similar.

Now let the total pressure on the low-pressure side be maintained. We consider the addition of protium to a system containing initially only tritium. Then the ratio of the tritium flux to its initial value is:

\[ J_T / J_{T_0} = y_T^{\text{total}} / y_T^{\text{total}} (y_T^{\text{total}})^{1/2} \]

whereas the ratio of the effective system tritium inventories under the same conditions is:

\[ C_T / C_{T_0} = \left( y_T^{\text{total}} / y_T^{\text{total}} \right)^{1/2} \]

\[ C_T / C_{T_0} = \left( y_T^{\text{total}} / y_T^{\text{total}} \right)^{1/2} \]

respectively, for a gas-phase and a liquid-metal phase constituting the principal tritium reservoir.

If the system tritium inventory is maintained constant, then, respectively, for the two cases:

\[ J_T / J_{T_0} = y_T^{1/2} \cdot 1, J_T / J_{T_0} = 1 \]

In the first case, although the system tritium inventory has not been changed, the tritium window area will have to be increased to provide tritium from the system at the same total rate. In the second case the isotopic change has not affected the required tritium window area. Thus, depending on what type of phase represents the principal tritium reservoir, the tritium window area required for a constant total rate of tritium recovery is either increased or not affected for a constant tritium inventory, but clearly is increased if the tritium inventory is to be reduced.

If, instead, the total hydrogen isotope pressure is maintained constant, then:

\[ J_T / J_{T_0} = y_T^{1/2} \]

in both cases, while respectively:

\[ C_T / C_{T_0} = y_T, C_T / C_{T_0} = y_T^{1/2} \]

Thus, in both cases, the tritium window area required to effect the tritium inventory reduction at a constant tritium inventory rate is increased by the factor 1/2, but the tritium inventory reduction is greater in the second case.

It is also clear that isotopic exchange will either enhance or not affect the performance of barriers to tritium migration for all the cases considered. Note also that the increase in tritium window area required for removal of tritium at a constant rate when no protium is added is 1/3 when the tritium inventory level is reduced by the factor \(1/2\) in either a gas-phase or a liquid-metal phase.
Isotopic Sampling Effects When Surface Processes are Rate-Limiting

Nickman(35) and Krone(36) examined another case of tritium recovery through a tritium window from a fluid stream that had been isotopically swamped with protium. This case involved the postulate that the permeation rate-limiting step was a slow surface process, the recombination of hydrogen isotope atoms followed by desorption from the low tritium partial-pressure side of the membrane. Adsorption-desorption processes, even if consideration is restricted to adsorption and desorption of hydrogen isotopes only, can be complex and there is a vast, often contradictory literature concerning this topic. Therefore, we can only briefly touch upon the subject. If the adsorption and desorption step on the upstream side of the barrier is rate limiting then a similar approach can be used in the analysis.

This particular step in the permeation process is carefully examined because there is evidence that, under practical thermonuclear reactor conditions, the recombination and desorption of tritium atoms may indeed be rate-limiting. For example, Goodill et al.(37) explained the results of their experiments concerning with permeation of hydrogen isotopes from liquid lithium through niobium membranes, on the basis of such a rate-limiting step.

The model for hydrogen isotope permeation which we shall consider involves the following assumptions, and approximations:

- All resistance in permeation other than those due to the desorption process are negligible.
- The partial pressures of tritium and protium on the low partial-pressure side of the barrier are so small that the adsorption rate on that side of the barrier is negligible in comparison to the desorption rate.
- The simple Langmuir adsorption theory with a single class of active sites adequately describes the adsorption process.
- Dissociation in the gas phase to form the atomic hydrogen isotope species from the diatomic molecular species can be neglected.

- The adsorptive characteristics of the high and low partial-pressure barrier surfaces do not differ significantly.
- The permeation occurs under steady-state, isothermal conditions.

The essence of the elementary Langmuir theory of adsorption may be stated as follows. Adsorption of molecules from the gas phase onto solid surface can occur only at certain active "sites" on the surface which have an affinity for the species being adsorbed, are uniformly distributed, and have equal affinities for the adsorbed species. Each such site is assumed to be capable of adsorbing only a single atom or molecule of the adsorbed species and, because the forces that are responsible for the adsorption are short range, the adsorption at any site is assumed to be independent of whether or not an adjacent site is occupied. If the adsorption involves a dissociation, say of a diatomic molecule, such as tritium molecules, to form the atomic species, then the adsorption process is assumed to involve a reaction between the diatomic molecule and two adjacent sites on the solid surface. The number of pairs of adjacent sites, based on the above listed characteristics of the active sites, is proportional to the square of the number of unoccupied sites, which can be represented in terms of a fractional occupancy of active sites by the adsorbed species, often called the surface coverage by the adsorbed species. In terms of the surface coverage by the adsorbed species and the partial pressure of the gas phase molecular form, a kinetically third-order expression for the rate of adsorption is postulated:

$$r_{\text{Ads}} = k_{\text{Ads}}(T)(1-\theta)^2P_{T_2}$$

Similarly, the desorption reaction is assumed to involve the kinetically second order reaction of two adsorbed atoms on adjacent active sites whose number is taken as proportional to the square of the fractional coverage and, in terms of surface coverage by the adsorbed species, a rate expression is written as:
Under conditions of adsorption equilibrium, the two rate expressions for adsorption and desorption can be equated and solved for $\theta_1$ to obtain:

$$\theta_1 = \frac{k_{T_2\text{Ads}}(T)}{k_{T_2\text{Des}}(T) P_{T_2}} \left[ \frac{1}{1 + \frac{k_{T_2\text{Ads}}(T)}{k_{T_2\text{Des}}(T) P_{T_2}}} \right]^{1/2}$$

Although other, more complex theories may describe the adsorption behavior in a particular system more accurately, the results obtained with the simple Langmuir theory should be representative and may be the most accurate in some cases, e.g., see Smith. Examples of other types of models for hydrogen isotope adsorption on, and desorption from metals are those of Bucur, et al., and Bucur for adsorption of protium on palladium films. They considered multiple types of active sites and surface reactions and there are many other such treatments. The general approach to analyzing of adsorption or desorption-step-limited permeation processes by using more complex adsorption and desorption models is similar.

The listed assumptions imply that the permeation flux, when only tritium is present, is given by the Langmuir expression for the rate of desorption of tritium from the downstream barrier surface. The surface coverage in the absence of significant upstream surface adsorption and bulk barrier metal diffusive resistances is that corresponding to equilibrium with the upstream molecular tritium partial pressure. The rate expression is therefore:

$$J_{T_2} = \frac{k_{T_2\text{Ads}}(T) P_{T_2}}{\left[ 1 + \frac{k_{T_2\text{Ads}}(T)}{k_{T_2\text{Des}}(T) P_{T_2}} \right]^{1/2}}$$

A similar analysis based on the same assumptions when both tritium and protium are present, can be summarized as follows. The rate expressions are of the form:

$$J_{T_2} = k_{T_2\text{Des}}(T) P_{T_2}^{1/2} \frac{1}{2} k_{HT\text{Des}}(T) P_{T_2}^{1/2}$$

and the absorption equilibrium expressions are of the form:

$$k_{T_2\text{Ads}}(T) P_{T_2}^{1/2} = k_{HT\text{Des}}(T) P_{HT}^{1/2}$$

The preceding adsorption equilibrium expression and the gas-phase isotopic interchange equilibrium expression can be solved for the diatomic protium and tritium and HT partial pressures in terms of the protium and tritium atom fractions in the gas phase and of the total hydrogen isotope partial pressure, although the solution may have to be obtained by numerical techniques, and the results inserted into the rate expressions. The presence of the third hydrogen isotope, deuterium, and significant dissociation of the molecular species to form the atomic species in the gas phase further complicate the problem, but the same principles are applicable.

If the tritium is isotopically swapped with protium, we have:

$$P_{H_2} \rightarrow P_{HT} \rightarrow P_{T_2} \rightarrow P_T \rightarrow P_H$$

and the rate and adsorption equilibrium expressions become, respectively:

$$J_{T_2} = \frac{1}{2} k_{HT\text{Des}}(T) P_{HT}^{1/2} \frac{1}{2} k_{HT\text{Des}}(T) P_{HT}^{1/2}$$

and:

$$\frac{1}{2} k_{T_2\text{Des}}(T) P_{HT}^{1/2} \frac{1}{2} k_{HT\text{Des}}(T) P_{HT}^{1/2}$$
Close to the root of a tree, or in a
thickly wooded area, the part of ground
covered by trees will be about equal to the
area of the tree crown. In such places, the
effect of evaporation will be much
reduced, and the surface will be
wetted for a longer time than in other
places.

which would increase the surface area, e.g., by
lowering the surface of the ground, or by the
action of a dense undergrowth of plants that
are able to use the moisture in the ground.
In experiments involving only a single hydrogen isotope.

Assume now that $\text{H}_2\text{O}$ and $\text{H}_2\text{O}$ form an ideal solution. Then, by definition:

$$\frac{\text{mole fraction of water}}{\text{mole fraction of water in liquid}} = \frac{\text{number of moles of water}}{\text{total number of moles}}$$

For a given liquid, the density $\rho$ and the specific gravity $\gamma$ of the liquid are related by:

$$\gamma = \frac{\rho}{\rho_0}$$

where $\rho_0$ is the density of water at 4°C. The specific gravity of the liquid can be used to determine its density.

The density of a liquid is typically measured at room temperature, which is often 20°C. Here are the densities of some common liquids at 20°C:

- Water: 1.0 g/cm$^3$
- Ethanol: 0.789 g/cm$^3$
- Isopropyl alcohol: 0.785 g/cm$^3$
- Hydrogen: 0.0899 g/cm$^3$
- Helium: 0.168 g/cm$^3$
- Neon: 0.801 g/cm$^3$

The specific gravity of a liquid is a measure of its density relative to water. The specific gravity of a liquid is an important property in many applications, such as in the chemical industry, where the density of a liquid is used to determine its concentration.
The graph illustrates the relationship between the variables. As the parameter increases, the function values also increase linearly. This is evident from the slope of the lines, which remain consistent throughout the graph. The data points are evenly distributed along the curve, indicating a strong correlation between the variables. The graph provides a visual representation of the theoretical predictions, confirming the expected outcomes under the given conditions.
If tritium recovery system performance is determined primarily by transport rates, then isotope pumping probably is not cost-effective and may be completely useless.

If tritium recovery system performance is determined primarily by equilibrium considerations, then isotope pumping may be cost effective, but there is no guarantee that this will be the case.

Isotope pumping will either not significantly affect or will enhance the performance of barriers to tritium escape into the biosphere for a specified tritium concentration level. The judgment of cost effectiveness must, of course, involve consideration of changes in, e.g., required recovery system capacity, requirements for isotope separation, benefits arising from reduction in tritium inventory, and in cases of escape of tritium into the biosphere, effects on other fusion reactor systems. Potential use of less expensive construction materials or lower energy consumption and costs of a unit of recovery system capacity.

Because of limitations imposed on the length of papers, we were unable to discuss the effects of the invariable presence of other hydrogen isotopes and of isotope pumping on other fusion reactor tritium recovery systems, and tritium migration mechanisms within and from fusion power plants. Examples of tritium recovery processes and migration mechanisms for which interesting analyses using similar methods can be performed include:

- the gas kinetic-limited permeation process discussed by Levin and Stickney
- the nonequilibrium permeation process described by Shuke and Stickney
- high-temperature lithium distillation for tritium recovery suggested by Pile and Wu
- the molten-salt extraction process for tritium recovery from liquid lithium described by Maroni, et al.
- permeation through potential nonmetallic materials of construction for fusion reactors, e.g., organic polymeric materials used for, e.g., gaskets and seals, and components, such as those discussed by Faucher, et al. on thermal oxidation or other corrosion layers that are potentially useful as tritium barriers, as discussed by Sjöholm and Savage
- thermal diffusion processes discussed by Pedersen (197).

REFERENCE