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# AN EVALUATION OF METHODS FOR RECOVERING TRITIUM FROM THE BLANKETS OR COOLANT SYSTEMS OF FUSION REACTORS

J. S. Watson

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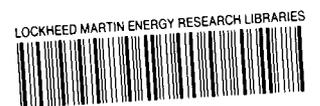
AN EVALUATION OF METHODS FOR RECOVERING TRITIUM FROM THE  
BLANKETS OR COOLANT SYSTEMS OF FUSION REACTORS

J. S. Watson

JULY 1972

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AN EVALUATION OF METHODS FOR RECOVERING TRITIUM FROM THE  
BLANKETS OR COOLANT SYSTEMS OF FUSION REACTORS

J. S. Watson

ABSTRACT

Potential methods for recovering tritium from the blankets or coolant systems of fusion reactors were reviewed and evaluated. The most restrictive requirement for the removal system will be prevention of an unacceptably large release of tritium to the environment. Diffusion of tritium through walls of heat exchangers into the steam turbine system is the most serious problem that must be solved since containment of tritium that reaches the steam system appears to be impractical. Based on estimates of future tritium release regulations, the study was made with a standard release rate of 4 Ci/day for a 2800-MW(e) system; however, the effects of other regulations, both more and less restrictive, were considered. The ORNL reference design featuring a potassium coolant and "topping" turbine coupled to a conventional steam turbine system was used as the basis for this study, but alternative systems that use a helium coolant and, possibly, gas turbines are discussed.

Two recovery methods appear to be promising. One is diffusion of the tritium through metal surfaces, probably palladium-coated niobium, into a slightly oxidizing atmosphere. By locating the metal surface in the high-temperature lithium system and coating the potassium--steam heat exchanger with a less permeable material such as tungsten, removal to the recovery system is strongly favored over loss to the steam system, and the required additional capital costs for tritium recovery appear to be acceptable.

The tritium can also be recovered by cold traps in the potassium system. However, unless a sorbent (e.g., yttrium) is used in the cold trap, it does not appear feasible to reduce the release rate to 4 Ci/day by this method; also, the cold traps would be too large to be economical. Traps filled with sorbent, however, do appear to be economical and promising. The development of more effective tritium barriers on the coolant-steam heat exchanger could alter this conclusion.

Neither gas sparging nor distillation appears to be an economical recovery process. At the low tritium concentrations required to prevent leakage of more than 4 Ci/day, binary distillation or sparging will not generate a lithium-free product.

A development program to further evaluate and demonstrate recovery methods is outlined. The emphasis should be directed at metal window systems and cold traps with sorbents. This program should be coordinated with power system design studies and with appropriate materials support work or consultations.

## 1. INTRODUCTION

This report presents the results of a survey of methods for recovering or removing tritium from the blanket of fusion reactors. This is one of two tritium handling problems likely to be significant in first-generation fusion reactors. The second problem, recovery and recycle of tritium from the main plasma fuel system, will be examined at a later date. Reprocessing plasma exhaust does not appear to be as difficult as recovery from the blanket or to raise as many questions of feasibility.

The purposes of this study were: (1) to define the magnitude of the tritium recovery problem and establish performance requirements for recovery systems, (2) to examine, in the light of these requirements, the several recovery systems that have been proposed and determine which of them are technically and economically feasible and which appear to be most promising, and (3) to suggest specific experimental data, which, at the present time, would be most likely to expedite the development and demonstration of economic recovery systems.

This study dealt primarily with the recovery of tritium from a blanket system proposed at ORNL and described by Fraas.<sup>1,2</sup> Other workers<sup>3,4</sup> have suggested other systems, the principal differences in which involve the coolant fluid and the maximum temperature allowed in the blanket. Most of the proposed systems use molten lithium in the blanket, but the operating temperatures vary between 600 and 1000°C. The prime ORNL system uses a molten potassium coolant between the lithium and the final steam system. Alternative proposals from ORNL and elsewhere would use a helium coolant instead of potassium. Some comparisons of the relative merits of helium and potassium have been given by Fraas.<sup>5</sup> When it appears appropriate, a comparison of the relative merits of potassium, helium, or alternative systems in permitting effective

tritium recovery is mentioned in this report. Some more advanced fusion power systems, which use direct energy conversion or magnetohydrodynamics, have also been proposed. However, the hardware required for these advanced concepts has not been sufficiently well defined that a study of tritium recovery problems is justified at this time.

The proposed potassium cooling system used for this study<sup>1,2</sup> is shown schematically in Fig. 1. Molten lithium is circulated through the blanket region surrounding the fusion chamber at the rate of 25,000 kg/sec. During its pass through the blanket, the lithium is heated from 1825°F to 1925°F. The lithium is then routed to a 75,000-ft<sup>2</sup> heat exchanger, sometimes called the potassium boiler. The potassium boiler will probably be constructed as part of the outer regions of the blanket system. The lithium is cooled to 1825°F in the exchanger by a potassium coolant. The 2500-kg/sec liquid potassium stream is heated from 1100°F, vaporized, and superheated to 1800°F. The potassium vapor then passes to a turbine generator that generates 1110 MW of power. Potassium leaving the turbine at 1100°C is in the form of vapor, which is condensed in a 160,000-ft<sup>2</sup> exchanger or steam generator and is recycled to the potassium boiler. The system produces 1050°F steam, which is fed to a conventional turbine-generator system and generates 1690 MW of power. The total power output from the system is 2800 MW; the thermal energy produced in the fusion core is 5000 MW. The unusually high thermal efficiency of this system results from the potassium turbine "topping system." The blanket system and all piping in contact with molten lithium are made of niobium. The potassium cooling system (other than the potassium boiler, which also contacts lithium) is made of stainless steel.

## 2. GROUND RULES

The minimum acceptable performance for a tritium removal system could be set by any of the following three requirements: (1) The inventory of tritium, and thus its concentration, in the blanket and heat transfer fluids must be low enough to provide an acceptably short fuel doubling time and to prevent excessive expenditure for tritium to start

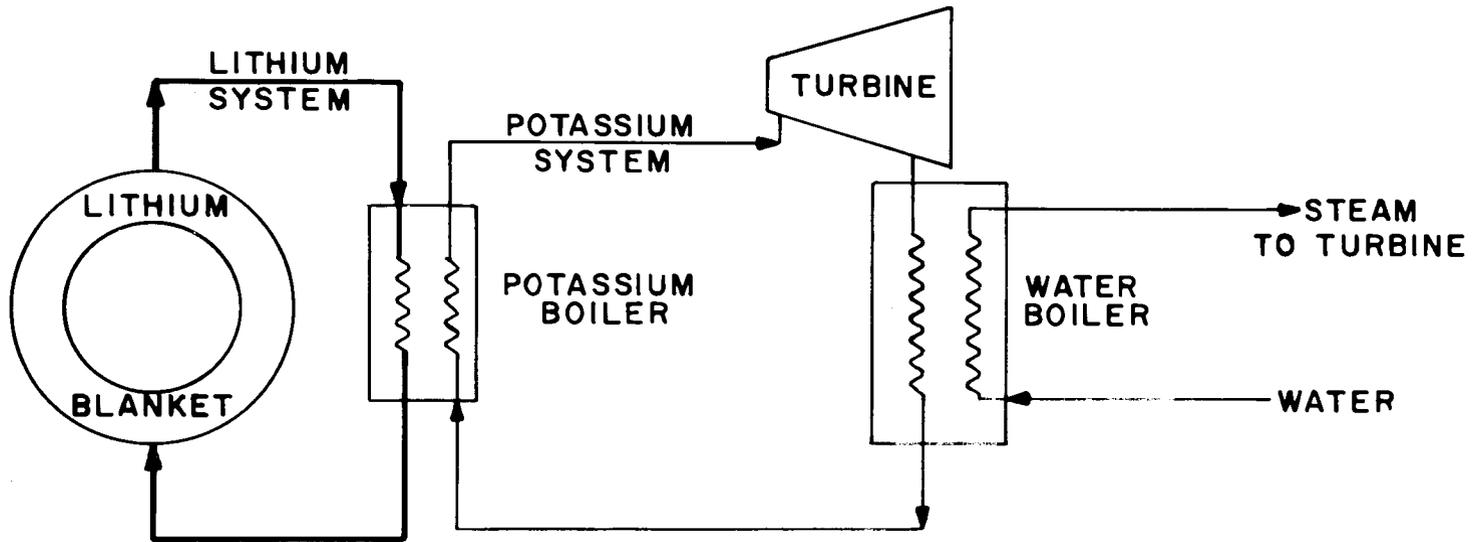


Fig. 1. Schematic of a Fusion Power Reactor Using Potassium Coolant.

the first generation of reactors. (2) The concentration of tritium must be maintained at a sufficiently low level to prevent unmanageable (hydrogen) embrittlement of structural materials in the blanket. Sorption of sufficient hydrogen or tritium by the niobium container will result in embrittlement when the metal is cooled. (3) The rate at which tritium is released to the environment must not exceed an acceptable limit. Since the release rate is a function of concentrations in the working fluids, this imposes limits on the allowable concentrations in these fluids. The third requirement imposes the most stringent conditions for the tritium removal system.

Maximum allowable release rates have not been set for fission reactors, and certainly there are no definite standards for fusion reactors. It is possible that separate and different tritium release standards will eventually be adopted for fission and fusion reactors since tritium is the only radioactive material released by fusion (other than isotopes produced by activation of structural materials). Fission systems, on the other hand, release at least small quantities of several other radioactive isotopes. However, in planning fusion systems, one should not expect or depend upon more lenient standards than those adopted for fission reactors.

The allowable release rate chosen for this study was 4 Ci/day for a 5000-MW(t) [or 2800-MW(e)] system. This release rate, selected<sup>6</sup> as a reasonable standard, is only an estimate of the degree of containment that will be expected of fusion systems. The tritium release is assumed to be at ground level with little dispersion. If the release is into a stack or tall cooling tower, somewhat larger releases may be permitted. Current trends suggest that release standards for all radioactive materials will become more restrictive in the future as additional reactor systems go into operation. With a single-step removal-containment system, costs are strongly related to the release rate; therefore, the effects of different release rates (other than the estimated standard) are considered in this study when they are significant.

Tritium can (and will) diffuse through all metal surfaces containing the blanket and cooling fluids. For this evaluation, we have looked at the leakage through only one part of this surface — the heat transfer

tubes in the potassium-water boiler. This boiler is a large heat exchanger and contains a significant portion of the metal surface separating the liquid-metal working fluids from the environment. One can envision methods for further containing the tritium that leaks from other piping into enclosed working spaces around the equipment. These spaces around the equipment could be sealed and evacuated by a pump discharging to a recovery system, or the atmosphere in these spaces could contain some low (but sufficient) concentrations of a "getter" material to reduce the tritium pressure to a negligible level. Very low, or even tracer, concentrations of oxygen (or possibly water) may be sufficient for this purpose. Furthermore, since the sealed walls enclosing the space around the equipment will be at lower temperatures, the permeability of the secondary walls around the equipment will be small.

Tritium that diffuses into the steam system through the heat transfer surfaces of the potassium-water boiler is assumed, in this study, to be lost to the environment. (It will quickly exchange with hydrogen in the steam and eventually be released to the environment.) Fraas<sup>2</sup> has discussed existing power plant experiences in reducing water leakage. Close control of water losses can be helpful in providing "backup" tritium retention (especially to impound tritium released via an accident or a malfunction of the primary removal system). However, if tritium release is limited only by water release rates, the extremely tight restrictions needed in the primary removal system are assumed in this study to be too expensive. Of course, the water system could be continuously processed to separate the tritium and hydrogen. A comparison of costs for water distillation and cryogenic hydrogen-tritium distillation has been made for the Molten Salt Reactor Program by students of the MIT Practice School.<sup>7</sup> Although the cryogenic system was shown to be the more economical of the two methods, this would be an expensive primary method for maintaining a suitably low release rate.

Thus, if diffusion through the water-potassium heat exchanger determines the release rate, the permeability of the potassium-water heat exchanger surface is an important parameter that affects the costs and performance of removal systems. This quantity, along with the allowable

release rate, determines the allowable activity (pressure) or concentration of tritium in the potassium and lithium systems. The higher the concentrations allowed, the less difficult and more expensive the removal system becomes (i.e., until a concentration that results in a significant embrittlement of niobium) is allowed.

The steam generator is to be constructed of stainless steel, which is highly permeable to tritium. Several different coatings that would drastically reduce the permeability of surfaces have been proposed. The most effective coatings may be oxides or other nonmetallic materials. A program is in progress in the Reactor Chemistry Division to study the feasibility of developing strong, stable, and reliable coatings for reducing tritium transport in molten-salt reactors.<sup>8</sup> The results obtained in the studies covered in this program will probably be useful to fusion systems as well. However, at the present time, reliable nonmetallic coatings that will reduce permeation rates by more than a factor of 10 have not been demonstrated. Fraas suggests that a 0.3-mm tungsten coating is feasible and reasonably effective. This coating is used as a reference surface for the study reported here, but the effects on removal system requirements of better barriers are also discussed.

Once an acceptable tritium release rate, the permeability of the potassium-water heat exchange surface, and the area of the heat exchanger are estimated, the maximum pressure of tritium in the potassium loop is fixed. From information on the concentration-pressure equilibrium, one can subsequently estimate the allowable tritium concentration in the potassium loop. With the tritium pressure in the potassium loop known, the area and the permeability of the lithium-potassium heat exchanger can be used to estimate the allowable pressure and composition in the lithium loop. The proposed removal systems are then constrained to keep the compositions of these streams at the calculated values or lower (e.g., to meet the estimated release standards).

The 5000-MW(t) reactor described by Fraas (and shown schematically in Fig. 1) uses a 160,000-ft<sup>2</sup> exchanger (water boiler) between the potassium and the steam systems. With a 0.3-mm tungsten coating on this surface and a permeability<sup>9</sup> (for tungsten at 600°C) of  $3.3 \times 10^{-5}$  cc (STP)-mm/hr-cm<sup>2</sup>-atm<sup>1/2</sup>, the pressure of tritium in the potassium system must be

less than  $1.45 \times 10^{-11}$  atm to maintain a leakage rate of less than 4 Ci/day (e.g., less than  $4 \times 10^{-4}$  g/day). This corresponds to a tritium concentration of approximately 0.003 ppm<sup>10</sup> in potassium. Fraas<sup>2</sup> suggests that up to 4 mole % lithium be added to the potassium to increase the solubility of tritium by a factor of approximately 10 (e.g., the concentration could be approximately 0.01 ppm). Both the concentrations and the pressure in the potassium are low by many standards, but it is believed that these are the most reasonable values to use at the present time. Improvements in tritium barriers or higher permissible release rates will increase the allowable pressure and concentration. On the other hand, a reduction in the permissible release rate would result in a corresponding lower pressure and concentration.

The tritium pressure in the lithium system is assumed to be only slightly higher than that in the potassium system if one chooses to remove essentially all of the tritium in the lithium loop. In this case, the total tritium transport through the 75,000-ft<sup>2</sup> potassium-lithium heat exchanger would be the same as the leakage rate,  $4 \times 10^{-4}$  g/day. Even if one coats the potassium-lithium heat exchanger with 0.3 mm of tungsten, the pressure would be only  $1.6 \times 10^{-11}$  atm. This relatively small increase in pressure results from the larger heat exchanger and the increased permeability of (even) tungsten at 1000°C. However, because of the greater affinity of lithium for tritium, the concentration of tritium will be substantially higher in the lithium system. The solubility of tritium in lithium at 1000°C and  $1.6 \times 10^{-11}$  atm can only be estimated by extrapolating data<sup>11</sup> to 1000°C. The tritium pressure could be higher since the temperature over the potassium boiler varies. The pressure on the potassium side may be higher at the higher-temperature end of the exchanger. A more exact calculation of lithium pressure will be made in the future. Vapor-liquid equilibrium and permeability data are needed over the temperature range. Of course, if the tritium is recovered from the potassium system instead of the lithium system, the tritium transport through the lithium-potassium exchanger would be much greater (i.e., essentially  $10^3$  g/day). This would result in considerably higher concentrations in the lithium systems. The higher concentration in that case, of course, would not aid in tritium removal.

With a helium coolant, several other options exist. If this coolant is used with a steam power system, the method of recovering tritium from the lithium stream is the same as that used with a potassium coolant. However, because the sizes of the heat exchanger surfaces would be different (i.e., larger), the maximum allowable pressure would be different (i.e., lower). In low-temperature systems (<600°C) where stainless steel is used both in the blanket and coolant, one could oxidize the tritium in the helium loop and either remove the tritium as T<sub>2</sub>O or simply use the reduced tritium pressure to further decrease the loss of tritium to the steam system. However, in high-temperature systems with a niobium heat exchanger separating the lithium and helium, corrosion will be severe unless the niobium is coated with a more resistant material.

If helium is used both as the coolant and as the turbine fluid, the problem will be considerably different. In this case, the helium stream would be cooled by water only after it reached a much lower temperature and pressure. The resulting large change in pressure and temperature would decrease the loss of tritium to the water coolant considerably, and helium leakage through pump and turbine seals would probably become the important consideration.

As noted earlier, these ground rules are based upon current estimates of release standards and upon current or near-current tritium barrier technology. In the latter area, the assumptions are probably (or hopefully) conservative; nevertheless, the release standards may be optimistic (from the view of the reactor operator). Since either of these parameters could change with time, an attempt has been made to cover a range of conditions around our best estimated values. We also want to introduce the idea of a completely effective barrier. This is a relatively expensive alternative which could be adopted if release rates are required to be considerably lower than those considered in this study. This would greatly simplify tritium recovery (tritium could be recovered at much higher overpressures and concentrations), although the barrier loop itself would be expensive and affect the entire heat removal-power generation system.

### 3. REMOVAL OF TRITIUM BY DIFFUSION THROUGH A METAL "WINDOW"

One method for removing tritium from the metal streams before it reaches the potassium-water heat exchanger would be to include an alternative escape route upstream. That is, a large permeable metal surface could be placed in either the lithium system or the potassium system to compete with the heat exchanger for tritium removal. This alternative escape surface is referred to as a "window" through which tritium is intentionally allowed to escape to a collection system. The window would probably consist of a shell-and-tube device having the appearance of a heat exchanger, although it would be operated isothermally. One could utilize the (upstream) position, a more permeable metal in the window, thinner tubes, and higher temperatures to favor diffusion through the window over diffusion through the heat exchanger.

#### 3.1 Location and Material

The most effective place to locate the window would be in the lithium system, upstream from the potassium boiler. Here, the tritium is at its highest pressure as well as its highest temperature. The permeability of metals increases with temperature. Another advantage of this position is that the resistance of the liquid metal film will be less in the lithium system than in the potassium system. Although palladium would be the most effective metal for the window, palladium is not compatible with lithium. Palladium at 1000°C has a permeability almost seven orders of magnitude higher than that of tungsten at 600°C.<sup>12</sup> Niobium is also an attractive material for the window. It has a permeability only about one-fifth that of palladium, but it is compatible with lithium and is less expensive. However, as noted later, niobium would require a palladium coating on one side to resist oxidation.

#### 3.2 Outside Pressure

The principal uncertainties in the "metal window" concept are the ability to produce the coated tubes required and the ability to reduce the partial pressure of tritium on the opposite face of the window to a

level significantly below  $1.6 \times 10^{-11}$  atm (e.g., below the pressure in the lithium stream). Such a low pressure, while possible to achieve, will be difficult to measure. One possible scheme would involve using a low concentration of oxygen in the recovery side of the window. This oxygen, although at a low concentration, would still be at a considerably higher pressure than the tritium. As the tritium diffused to the outer surface of the window, it would emerge on a catalytic surface (palladium) in the presence of a significant excess of oxygen. This should reduce the tritium pressure to the dissociation pressure of water at that temperature and composition (water and oxygen). Tritium pressures of less than  $10^{-11}$  atm may be possible at  $1000^{\circ}\text{C}$  with an oxygen pressure of only a few thousandths of an atmosphere; even less oxygen may be required at lower temperatures.

The tritium could then be recovered by regeneratively cooling a side stream of the recovery gas ( $\text{He} + \text{O}_2 + \text{T}_2\text{O}$ ), sorbing the  $\text{T}_2\text{O}$  on a desiccant, and periodically regenerating the desiccant (with heat) to recover the  $\text{T}_2\text{O}$ , which would be caught on condensers. A small (e.g., 1-kg  $\text{T}_2$ /day or 6-kg  $\text{T}_2\text{O}$ /day) electrolysis system would be required to reconvert the tritium to the elemental state for recycle to the fuel system.

While the tritium is in the  $\text{T}_2\text{O}$  state, an essentially negligible loss through the system walls is anticipated. After reversion to the elemental state, the loss would remain low due to the low temperatures; however, the atmosphere around the equipment would need to be contained and sealed as with any tritium handling system.

This system would require a permeable, oxygen-resistant coating on the niobium tubes. Palladium is an obvious contender for this service, but the ability to make reliable coated tubes has not been demonstrated. This leaves an uncertainty in the feasibility of this process.

### 3.3 Resistance of the Liquid Metal Film

The effectiveness of the metal-window removal system depends upon the differences in tritium permeability for various metals. If the mass transfer rate should be significantly affected by the flow resistances in either side of the metal window, the process would lose some of its

advantage. If film resistance controlled the transfer rate, there could be no difference between niobium-palladium and tungsten windows; and, under the ground rules of this study, metal windows would not be practical removal systems.

The film resistance in the recovery system (e.g., the He-O<sub>2</sub>-T<sub>2</sub>O stream) is expected to be essentially zero. As noted earlier, the oxygen pressure in this stream will be considerably higher than the tritium pressure, even in the metal loop. Thus, a substantial excess of oxygen will be available, and oxygen transport to the metal surface will not hinder the transfer rate. Similarly, the slight elevation in T<sub>2</sub>O concentration (like the slight depression of O<sub>2</sub> concentration) at the metal surface is not expected to shift equilibria at the interface sufficiently to create a significant tritium concentration at the metal-gas interface.

As noted earlier, the most desirable location for the metal window is in the lithium system. To estimate the film resistance in the lithium system, a 1-in.-ID pipe and a liquid velocity of 2 fps were selected as typical conditions. With lithium metal at 1000°C, these conditions correspond to a Reynolds number of approximately 50,000. The diffusion coefficient of tritium in lithium was assumed to be approximately  $4 \times 10^{-5}$  cm<sup>2</sup>/sec.<sup>9</sup> (There are no measured values for this quantity; however, this is believed to be a reasonable guess.) The mass transfer coefficient was then estimated to be 0.0092 cm/sec, using the Dittu-Boelter equation (see Appendix 1 for details). The flux of tritium through the metal-metal window will be approximately  $1.29 \times 10^{-10}$  g-moles/cm<sup>2</sup>-sec or less (see Sect. 3.4), which corresponds to a concentration drop across the liquid-metal film of  $1.41 \times 10^{-8}$  g-moles/cm<sup>3</sup>. The concentration of tritium in the lithium can be estimated only by extrapolating the existing data to considerably higher temperatures (see Appendix B). Experimental data were obtained for temperatures up to only 825°C,<sup>10</sup> but an attempt was made to extrapolate those data to the higher temperatures of current interest. The resulting estimate predicted a metal composition (in equilibrium with a tritium pressure of  $1.6 \times 10^{-11}$  atm) of  $2.8 \times 10^{-7}$  g-atom/cm<sup>3</sup>. Since the concentration drop across the metal film ( $1.41 \times 10^{-8}$  g-moles/cm<sup>3</sup>) is a fraction of the bulk metal concentration ( $2.8 \times 10^{-7}$  g-moles/cm<sup>3</sup>), the concentration of the liquid metal at the

surface of the window is not greatly reduced by film resistance. This is an encouraging observation, although it is admittedly based upon assumed values for several parameters. Furthermore, a twofold or greater error in the film resistance, possibly due to inaccuracies in the assumed diffusion coefficient, could make film resistance significant. This result also shows that the window area will not be reduced more than a few-fold by using thinner tubes. The palladium cost, however, can be reduced considerably by the use of thinner tubes,

If the window were located in the potassium system, it could be placed between the lithium-potassium heat exchanger and the potassium turbine. In this case, the working fluid would be in the vapor state, and the metal window would have a higher temperature than if it were located in the liquid potassium system between the water boiler and the potassium boiler.

Let us consider a window located in the liquid potassium region. The estimated tritium mass transfer coefficient in liquid potassium is close to that estimated for lithium (see Appendix A). Some of the similarity results because the same value was assumed for the diffusion coefficient, although the temperature in the liquid potassium regions is substantially lower (600°C). The assumed diffusion coefficient is probably more conservative at the higher temperature, and less conservative at the lower temperature. The concentration of tritium in the potassium system will be considerably less than that in the lithium system (this situation could be improved by adding up to 4 at. % lithium to the potassium; however, the flux is also less, largely because of the lower temperature. With an average tritium flux of  $5.74 \times 10^{-11}$  g-atom/cm<sup>2</sup>-sec through the window, the concentration drop across the liquid film would be  $7.3 \times 10^{-9}$  g-atom/cm<sup>3</sup> and the minimum concentration in the bulk potassium system (e.g., just downstream from the window) would be  $1.4 \times 10^{-9}$  g-atom/cm<sup>3</sup>. Thus, liquid film resistance is significant and will probably control the size of the window. Even if 4 at. % lithium is added to the potassium, film resistance will be important. It is obvious, then, that this is not the best place to locate the metal window.

As noted earlier, another possible location for the window is in the potassium system between the potassium boiler and the potassium turbine. Here, one can take advantage of the higher temperature. Also, the tritium-to-potassium mole ratio will be slightly higher (than in the liquid potassium system) because additional tritium enters the potassium stream in the potassium boiler. On the other hand, the concentration of tritium will be lower due to the lower density of the vapor; however, this may be partially compensated by the higher diffusion coefficient in the gas. Using the Gilliland correlation,<sup>13</sup> the diffusion coefficient of  $T_2$  in potassium vapor at 1000°C is estimated to be  $0.467 \text{ cm}^2/\text{sec}$ . The potassium vapor is assumed to be an ideal gas. Again, a 1-in.-ID tube was selected for a typical or reference system, but a velocity of 90 fps was selected for the gas flow. These conditions give a mass transfer coefficient of 4.1 cm/sec (see Appendix A) and a (maximum) bulk concentration of  $5 \times 10^{-15} \text{ g-moles/cm}^3$ . The mass transfer rate in this case is controlled by film resistance, and the required window area is not competitive with a window located in the lithium loop.

### 3.4 Areas of Metal Windows Required

If we conclude that liquid or gas film resistance does not significantly affect transfer rates in the lithium loop, estimation of the required metal window size becomes relatively simple since only the resistance in the metal wall needs to be taken into account. Although Nb-Pd tubes will be required, the cost estimates given here will be based upon the use of palladium. This will add some conservatism in costs to offset the cost of preparing coated (two nickel) tubes. Actual tube costs may be lower if the tubes are made mostly of niobium, with little palladium. The cost estimate will be crude, but it will indicate if the process is obviously uneconomical. A tube thickness of 1/3 mm (84 mils) was assumed. The assumed tube thickness is somewhat greater than that used in conventional boiler tubes and may be conservative. Yet, the strength and reliability of palladium-niobium tubes are not likely to match those of conventional steel tubes; consequently, some additional wall thickness appears justified at the present time. An

optimization of tube diameter, thickness, etc., will be needed, but the assumed tube dimensions are believed to be sufficient for the immediate needs of this study.

The permeability of palladium was estimated by extrapolating data reported by Webb<sup>12</sup> and plotted by Fraas.<sup>2</sup> The permeability was estimated to be 900 [cc (STP)-mm/min-cm<sup>2</sup> · atm<sup>1/2</sup>] at 1000°C and 300 [cc (STP)-mm/min-cm<sup>2</sup> · atm<sup>1/2</sup>] at 600°C. The area required to remove 1 kg of tritium from the lithium stream is a function of the permeability of the water boiler and the allowed release rate, as discussed previously in Sect. 2. The required area is shown in Table 1. At the reference conditions (loss of 4 Ci of tritium per day through 0.3 mm of tungsten, e.g., partial pressure of tritium of  $1.5 \times 10^{-11}$  atm), the required area would be 16,000 ft<sup>2</sup>.

If the window were located in the potassium vapor or liquid regions, the area would be considerably larger due to the lower temperatures and film resistance.

Costs of metal windows will be approximately proportional to the window areas because of the strong contribution of the cost of palladium (actually Pd-Nb) metal to the total cost. With the 1/3-mm wall thickness selected for this study and a cost of \$38/oz for palladium, the cost of the unfabricated palladium metal will amount to \$10.8/ft<sup>2</sup>. The cost of preparing Nb-Pd tubes (if reliable tubes can be made) is unknown; we will assume that the total cost would be approximately the same as for palladium. It will also be necessary to construct the shell (probably of stainless steel), and to prepare a relatively small water removal and T<sub>2</sub> production system. This system would probably consist of a regenerative heat exchanger, a pair of desiccant beds, a condenser, and an electrolytic T<sub>2</sub> generator. It would be relatively small and will not be assumed to be a major cost item. Fabrication of tubes and construction of a stainless steel shell were estimated to increase the cost by an amount approximately equal to the total cost of an equivalent-sized stainless steel heat exchanger plus the additional inspections required for nuclear applications. The cost of stainless steel heat exchangers in 1967, as reported by Peters and Timmerhaus,<sup>14</sup> was increased by a factor of 1.28 to account for inflation, or an annual cost increase rate of approximately 5% to adjust prices to a 1972 basis. The resulting cost was then

Table 1. Estimated Cost of Metal Window

Partial Pressure, $P_1$ of $T_2$ (atm)	Window Area		Cost of Palladium (dollars)	Cost of Window Shell (dollars)	Total Cost of Window (dollars)
	(cm <sup>2</sup> )	(ft <sup>2</sup> )			
$5.0 \times 10^{-12}$	$25.8 \times 10^6$	27,734	299,523	367,536	667,060
$1.0 \times 10^{-11}$	$18.2 \times 10^6$	$1.96 \times 10^4$	212,000	284,000	495,000
$1.5 \times 10^{-11}$	$14.9 \times 10^6$	16,012	173,000	244,000	417,000
$2.0 \times 10^{-11}$	$12.9 \times 10^6$	13,867	149,762	219,421	369,000
$4.0 \times 10^{-11}$	$9.1 \times 10^6$	9,805	105,900	169,500	275,000
$10^{-10}$	$5.76 \times 10^6$	6,201	67,000	120,560	187,533
$2.0 \times 10^{-10}$	$4.07 \times 10^6$	4,385	47,360	93,150	140,500
$4.0 \times 10^{-10}$	$2.88 \times 10^6$	3,101	33,490	71,974	105,500
$10^{-9}$	$1.82 \times 10^6$	1,961	21,180	51,180	72,400

doubled to account for additional inspection costs required in most nuclear applications.<sup>15</sup> The resulting "major equipment" costs are shown in Fig. 2 for the cases considered in this study.

These numbers are useful for crude estimates or comparisons of the costs of removal; however, there are many other costs such as those for piping, electrical services, instrumentation, thermal insulation, buildings, land, overhead costs, etc., which would require much more detailed information to itemize. In many chemical plants, these costs are often essentially proportional to the "major equipment" cost, and the total cost will be four to five times the cost for the major equipment. A factor of 4 or 5 is probably reasonable to apply in our case. The high cost of tubes in the major equipment may suggest a lower factor, but more stringent building containment requirements, instrumentation, etc., would compensate for some or most of this advantage. Comparison of the major equipment costs is a useful way of comparing the relative costs for the various removal methods since the total costs will be approximately proportional to these values.

### 3.5 Principal Uncertainties and Possible Areas for Improvement

The principal uncertainties relative to feasibility of the metal window concept are the ability to produce reliable Nb-Pd tubes and the ability to maintain the tritium pressure in the He-O<sub>2</sub> stream at a sufficiently low value. For the reference system, this pressure is approximately  $10^{-12}$  atm. Better barriers would allow a higher pressure, but more strict tritium release standards would lower it. The permeation-catalysis system has been demonstrated to be "effective" in removing hydrogen (and hence tritium) from systems of low partial pressures.<sup>16</sup> The uncertainty exists only because the unusually low pressures have not been demonstrated; however, pressures close to those needed have been reached, and there is no indication that lower values cannot be reached. Estimates of the reliability or lifetime of two metal tubes are beyond the scope of this study; these will require experimental demonstration. Many factors such as tube fabrication, catalyst poisoning, corrosion, etc.,

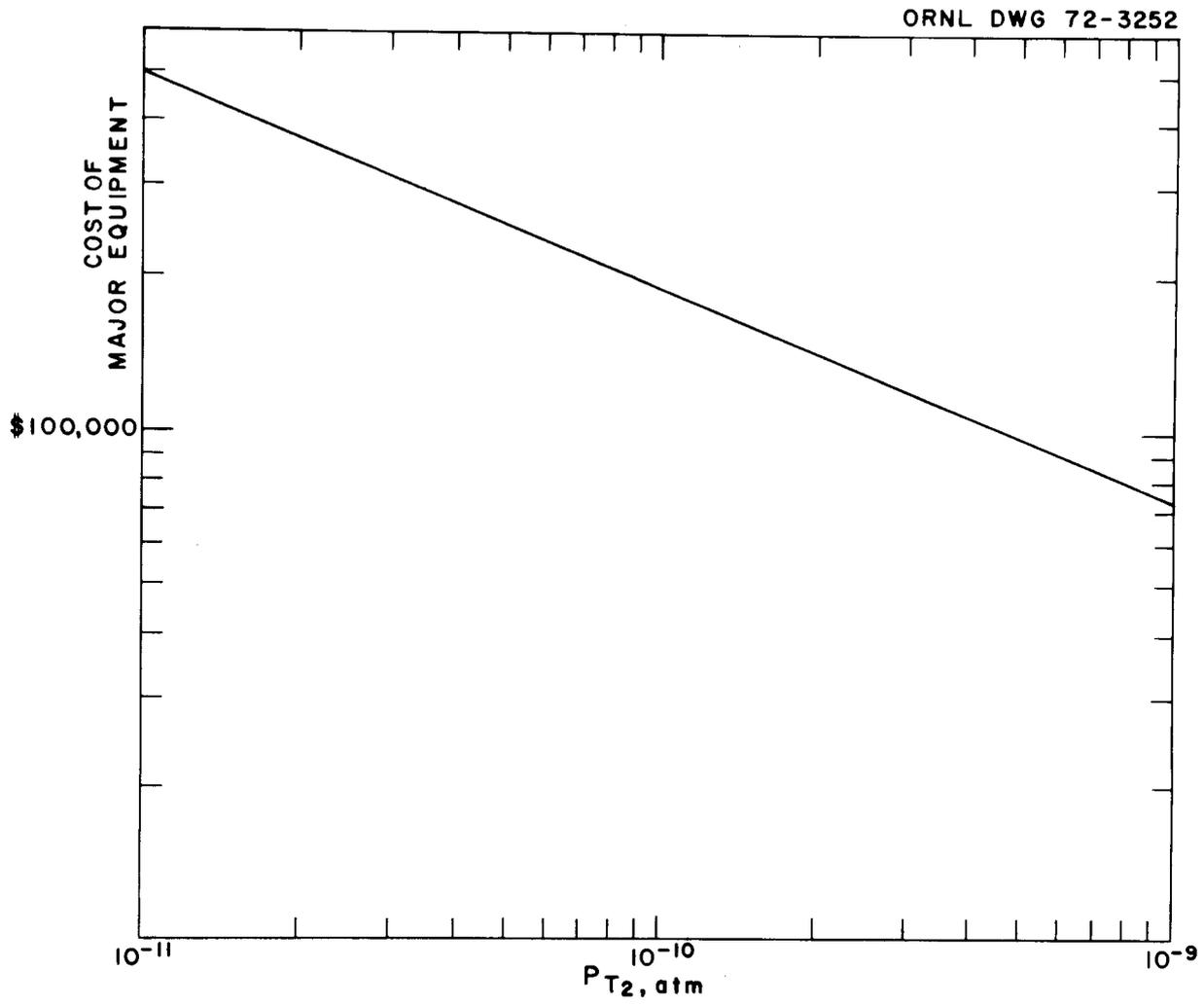


Fig. 2. Estimated Cost of Metal Window and Shell.

could make metal windows impractical; however, these are not necessarily fundamental problems, and, in the event that difficulties develop, a solution may be possible. Another significant uncertainty in the window concept concerns film resistance.

The economics of metal windows would be improved considerably if less palladium could be used. Niobium has a permeability only one-fifth that of palladium, but it is considerably cheaper (a factor of 20). Although essentially unaffected by liquid lithium, niobium is rapidly attacked by oxygen at high temperatures; so a niobium tube will require at least a thin palladium coating on the He-O<sub>2</sub> side. The thickness of this coating will affect the economics.

#### 4. COLD TRAPS

If one of the liquid metals (probably the potassium) is cooled sufficiently, the metal hydride will precipitate. The solid may be trapped on a filter or some other collecting surface which can be periodically reheated to release the tritium by decomposing the hydride at a higher temperature. This "cold trap" removal scheme was proposed and described by Fraas.<sup>2</sup>

A simplified sketch of the possible removal system is shown in Fig. 3. A process stream is removed from the power generation system probably at the lowest temperature point (e.g., from the liquid potassium between the water boiler and the potassium boiler). The process stream is cooled in a regenerative heat exchanger to minimize the net heat loss from the power system. The last few degrees of cooling will be effected by a separate cooler rejecting heat to an external source (possibly to preheat boiler feed water). The cooling achieved in this external heat exchanger determines the temperature drop across the regenerative metal-metal heat exchanger, and, in turn, the size of the exchanger. The less the external cooling, the less the thermal efficiency of the power system is impaired but the greater the regenerator surface area required. Some optimization is needed. Fraas suggests a 50°F external cooling temperature (or temperature drop across the regenerator). We considered this to be a reasonable value, and used it

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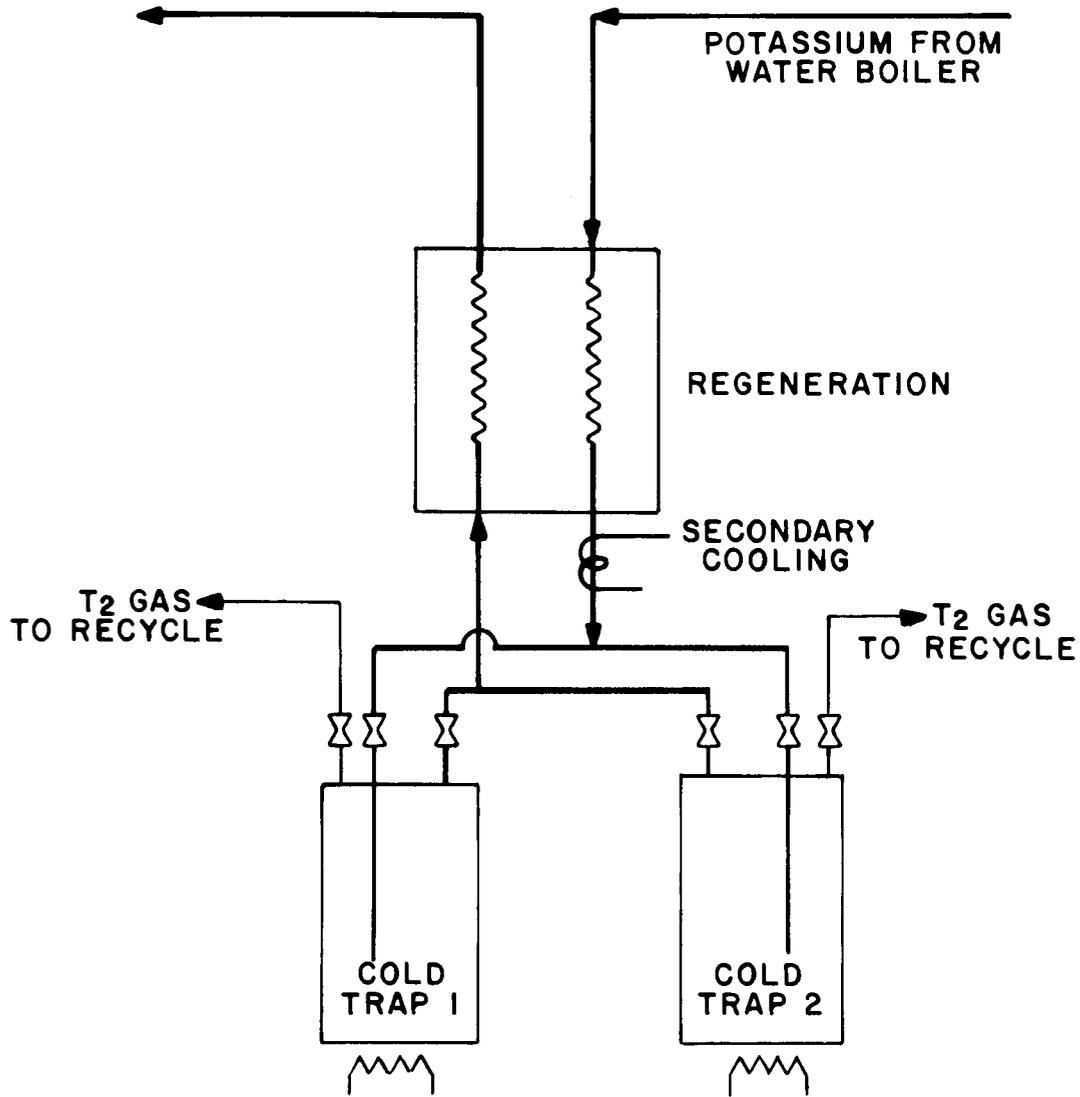


Fig. 3. Schematic of a Cold Trap System.

in this study. The cold trap itself would probably consist of a vessel containing a large solid surface area, possibly a mesh packing. The cold trap would be similar to oxide precipitation cold traps used with liquid-metal-cooled fission reactors.

To evaluate the cold trap concept, several aspects of the system were checked. These included solubility limits, size of the regenerator, loss of tritium through the regenerator walls to the exit stream, and size of the cold trap.

#### 4.1 Solubility Limitations

Fraas proposed cooling the potassium stream to 180°F (82.2°C). This is a moderately safe temperature increment above the freezing point of potassium (144.5°F or 62°C). The concentration of tritium allowed in the potassium loop was reported in Sect. 2 to be 0.003 wt ppm if the tritium release rate is 4 Ci/day and the water boiler contains a 0.3-mm layer of tungsten. The limit would be approximately 0.01 wt ppm if the potassium contains 4% lithium. This latter value is essentially the same as the solubility estimated by Fraas (data from p. 167 of Ref. 10) at 180°F. Thus, the entire potassium flow would have to pass through the cold trap to reduce the concentration to the level desired by the ground rules of this study. Even this performance is not adequate since the cold trap is located upstream from the potassium boiler. During the pass through the potassium boiler, the potassium picks up additional tritium equivalent to 0.0046 ppm; thus the concentration at the critical point in the system (e.g., at the boiler) would be greater than the specified limit. Location of the cold trap between the potassium boiler and the water boiler (e.g., in a potassium vapor region) is not attractive since regenerators are not effective when a change of phase is involved.

The solubility limitations just described should not rule out cold traps completely. The ground rules for this study do not represent fixed limits. As noted earlier, these rules could be altered by improvements in tritium barriers or by adoption of release regulations different from those assumed in this study.

#### 4.1.1 Size and Cost of the Regenerator

No detailed or optimized regenerator design was made. The objective of this study was simply to determine whether a practical regenerator was possible and to roughly estimate its cost. A metal velocity of 2 fps and a 1-in. hydraulic diameter (or both sides of the regenerator) were assumed. This corresponds to an average Reynolds number of about 50,000. The Reynolds number varies with temperature because of the variation in viscosity. The average Prandtl number was assumed to be 0.00264. Using the Lyon equation for constant heat flux, the inside and outside coefficients were estimated to be  $0.5 \text{ cal/cm}^2\text{-sec-}^\circ\text{C}$ . A 1/3-mm stainless steel wall (85 mils) gives an overall heat transfer coefficient of  $0.21 \text{ cal/cm}^2\text{-sec-}^\circ\text{C}$ . The area of exchange surface required for a  $50^\circ\text{F}$  ( $27.8^\circ\text{C}$ ) temperature drop is  $1.42 \times 10^7 \text{ cm}^2$  or  $15,000 \text{ ft}^2$ . This is a reasonable size; that is, the cost of the regenerator should not restrict the application of cold traps. Peters and Timmerhaus have reported<sup>14</sup> that stainless steel heat exchangers cost  $\$0.45/\text{ft}^2$  in 1967. Assuming a 5% annual increase in cost from 1967 to 1972 and doubling the cost<sup>15</sup> to account for extra inspections required for a "nuclear" application, the capital cost of the regenerator amounts to approximately  $\$17,000$ .

#### 4.2 Loss of Tritium in the Regenerator

Another potential problem associated with cold traps is the leakage of tritium across the walls of the regenerative heat exchanger. In the regenerator, a metal with a relatively high tritium concentration (the inlet metal) is separated from a metal at low concentration (the exit metal) by a thin metal wall. "Leakage" through this wall will add tritium to the "purified" stream and hamper the performance of the cold trap system. We need to determine whether this leakage is significant.

The leakage through a regenerator is calculated in Appendix D. For simplicity, the cold trap vessel (not the overall system) was assumed to be completely effective (i.e., that all of the tritium entering the cold trap vessel remains there). The potassium entering the regenerator from the cold trap is thus assumed to be free of tritium. The permeability

of the metal heat exchanger wall is a function of temperature, increasing as the temperature increases. Material balances show that the concentration difference across the metal remains constant along the path of the regenerator; therefore, the leakage rate varies with temperature along this path. The leakage rate is highest at the hot end of the regenerator and lowest (probably negligible) at the cold end.

The permeability constant is defined by:

$$K_{ss} = \frac{\phi \delta}{p_1^{1/2} - p_2^{1/2}},$$

where  $\phi$  is the flux of tritium through the metal exchanger wall,  $\delta$  is the wall thickness,  $p_1$  and  $p_2$  represent the tritium pressure on each side of the wall, and  $K_{ss}$  is the permeability constant of the stainless steel wall. The permeability constant is assumed to change with temperature in the following manner:

$$K_{ss} = A_1 \exp(-A_2/T),$$

where  $A_1$  and  $A_2$  are constants that are dependent on the wall material and  $T$  is the absolute temperature of the wall. The concentration of the tritium in the potassium or potassium-lithium mixture is proportional to the square root of the pressure:

$$C = K_p \sqrt{P}.$$

The constant  $K_p$  will also change (increase) with decreasing temperature. However, since the decrease of  $K_{ss}$  alone with temperature prevented a significant loss through the regenerator walls, the effect of temperature on  $K_p$  was not taken into account. The high-temperature value was assumed through the regenerator. This is a conservative assumption since the real loss will be less than that calculated.

A material balance gives the following expression for the concentration along the regenerator flow path:

$$-\frac{dC_1}{dz} = \frac{dC_2}{dz} = \frac{\phi A}{V} = \frac{AA_1 \exp(-A_2/T)}{K_p V \delta} (C_1 - C_2),$$

where  $Z$  is the distance along the flow path,  $A$  is the regenerator surface area per unit length, and  $V$  is the metal velocity. (This will be approximately the same in both inlet and outlet flow channels.) The difference in concentration across the regenerator tube wall ( $C_1 - C_2$ ) is a constant along the flow path. This constant is initially unknown; it is the principal parameter to be determined. At the entrance to the regenerator,  $C_1$  is the inlet concentration, a fixed parameter. Across the metal tube wall is the outlet concentration,  $C_2$ ; and the difference between  $C_2$  and the solubility at the cold trap temperature indicates the leakage. Integrating the material balance equation and solving for the constant ( $C_1 - C_2$ ), one finds that, at the conditions of interest, leakage of tritium through the regenerator will not be a major problem in cold trap design. Details of these calculations are given in Appendix D. The leakage, however, is a function of several variables, and one could propose a system where it would become significant. A larger regenerator surface area or a higher initial temperature would increase the loss.

#### 4.3 Size of the Cold Trap

The major shortcoming of any cold trap system is probably the large size required for the cold trap vessel. A large vessel is expensive to build, and it incurs large potassium and tritium inventory costs. No reference to tritium-potassium or hydrogen-potassium cold trapping rates (or required vessel size) has been found in the literature. On the other hand, considerable data and experience are available on cold trapping of oxides from liquid sodium. These data were obtained in programs connected with the liquid-metal-cooled fast breeder (fission) reactor. The oxide and hydride systems could differ in several respects that would make the oxide system a poor model for a hydride system; however, this is the best model available at the present time.

Two relatively recent review articles have summarized the experience with cold traps in large sodium systems.<sup>17,18</sup> For small units, natural-circulation cold traps have been used successfully; for large units, forced-circulation units with regenerators are used. The cold trap is usually a vessel filled with a mesh or other type of packing, which acts either as a filter for the solid precipitate or as a solid surface on which the precipitate forms. Various designs use different packings and liquid flow paths. A considerable quantity of oxide can be removed in a cold trap, and plugging can cause difficulties. To reduce tritium inventories, a tritium cold trap would be cycled (e.g., reheated) frequently; with this treatment, plugging would probably not cause a severe upset unless a buildup of impurities (possibly oxides) occurred.

Cold traps are sized to provide approximately 5 min of residence time in the cold trap vessel. This practice is based upon experimental studies by Bruggeman.<sup>19</sup> If the entire potassium stream from a fusion system passed through the cold trap, each trap (two traps are required for continuous operation) would contain 750,000 kg or 33,000 ft<sup>3</sup> of potassium to provide 5 min of residence time. If only a fraction of the potassium is directed through the cold trap, then the required volume would be reduced by that fraction. Unless the diverted streams can be reduced to only a very small fraction, the cold traps will be large and expensive, and potassium (and thus tritium) inventories will be increased considerably by the cold traps. Potassium costs approximately \$0.60/kg. If 4 at. % lithium is used, the cost would be further increased to approximately \$1.3/kg. This corresponds to a cost of approximately \$2 million for two traps if the entire potassium stream flows through the trap. York mesh packing in large quantities costs \$30 to \$36/ft<sup>3</sup>,<sup>20</sup> or an additional \$2 million for two traps to handle the entire potassium flow. Since inventory considerations will not permit the buildup of tritium to high concentrations, efficient tritium removal by simply heating such large volumes of metal may also be a problem.

#### 4.4 Uncertainties and Areas for Improvement in Cold Trap Performance

The solubility and pressure-concentration relations for the cold traps are estimated from data obtained with similar systems; however, such data are not particularly reliable. Until quantitative measurements are made on K-Li-T systems, the solubility limitations assumed in this study should be considered as only tentative estimates. These parameters are of interest since the equilibrium conditions determine the maximum performance of cold traps.

The principal uncertainty in cold trapping is the required size of the trap. If 5 min is the required residence time (as discussed previously), cold traps will probably not be economical for tritium removal unless a more effective barrier is used between the potassium and the steam. Improvements in the barrier will allow a reduction in potassium flow rate through the cold trap and, hence, a reduction in cold trap size.

There is reason to hope that somewhat more effective cold trap designs can be developed in the future. The required residence time is determined by crystallization characteristics of the hydride, which could be different from those of oxides. Results from a recent study by McPheeters and Biery<sup>21,22</sup> suggest that the oxide removal rate is proportional to the internal surface area of the trap, indicating that the solid oxide forms on the metal surface and its presence is not due to filtering action. If plugging does not cause problems with hydrides, perhaps a substantially greater surface area per unit volume can be included in hydride cold traps; this would reduce their required size.

#### 5. SOLID SORBENTS

Solid sorbents have also been proposed for tritium removal. The most likely candidate at present appears to be yttrium metal. Yttrium forms a strong metal hydride, which is stable over the temperature range of interest for this study. From an equilibrium point of view, the use of a solid sorbent would be superior to a simple cold trap system. Again,

however, some difficulty will result from the very low pressures and concentrations involved. Without a detailed investigation of mass transfer rates, column design, and fraction of approach to equilibrium loading, we will simply consider a perfect system in which all the sorbent comes to equilibrium with the liquid metal (or with the vapor over the metal).

In order to obtain reasonable equilibrium loadings, the liquid metal will have to be cooled considerably. This can be done without a large heat loss if a regenerative heat exchanger is used. Thus, the sorber system would resemble a cold trap system. The principal difference is that the vessel would be filled with a sorbent (yttrium) rather than mesh. Regeneration of the sorbent would be achieved by periodically heating the sorber bed. For continuous operation, at least two separate beds would be required.

The most recent study of the yttrium-hydrogen (tritium) system reviewed for this study was that of Yannopoulos, Edwards, and Wahlbeck.<sup>23</sup> Their data did not extend below 6 at. % tritium or to temperatures below 651°C. The pressure of tritium (assuming it to be approximately the same as hydrogen) remains far above the desired values at their conditions; consequently, it is necessary to extrapolate the results to lower temperatures and lower atom fractions of tritium. The lowest (composition) data point at each temperature of Yannopoulos' study was taken as a reference point. Extrapolation to lower compositions was assumed to follow Sievert's law; that is,

$$X_T = k \sqrt{P_{T_2}} .$$

The value of  $k$  was assumed to vary with temperature in accordance with the expression

$$\ln k \propto \frac{1}{T} .$$

The data of Yannopoulos fit this expression well (see Appendix C). One can reduce the equilibrium pressure by decreasing either the temperature or the atom fraction loading,  $X$ . The combinations of  $T$  and  $X$  that will give a pressure of  $1.45 \times 10^{-11}$  atm ( $5 \times 10^{-9}$  mm) are given in Appendix B.

These equilibrium values give only the maximum loading. Because of mass transfer limitations, the overall loading of the packed bed would be less. If the bed were cooled below 82°C (180°F), the maximum loading would be 0.05 atom fraction. The price of yttrium is approximately \$150/lb; hence, the cost of yttrium metal in each bed would be approximately \$120,000 if the beds were regenerated daily. Of course, the cost of yttrium metal would be proportional to the length of time allowed between regenerations. Larger beds would, however, allow larger portions of the bed to approach the equilibrium loading. Yttrium is an expensive sorbent. Lanthanum and cerium, which should behave similarly, may cost as little as \$40/lb. Zirconium (possibly even with traces of hafnium) may prove to be a more economical sorbent.

The use of sorbents can be considered a modification of the cold trap concept. With solid sorbents, higher temperatures could be used and, in turn, the size of the regenerator and the heat loss would be less than with cold traps. Solid sorbents also eliminate the absoluteness of the equilibrium or solubility limitations. It is also possible that they may reduce the required vessel volume. Since tritium rather than tritide molecules will be diffusing to the solid (sorbent) surface, one might expect mass transfer rates to be higher in a sorber than in an empty cold trap. Use of a solid sorbent would require purchase and maintenance of the sorbent bed, and higher pressure drops probably would be experienced than with cold traps. The lifetime of the sorbent (e.g., number of metal-to-tritide-to-metal cycles permitted before severe degradation of the sorbent occurs) is unknown. The problems of recovering tritium from the bed would be similar to those of recovery from cold traps. The accumulation of other impurities, especially oxides, may also cause difficulties with sorber beds.

There is perhaps less known about solid sorbent beds than about any of the processes reviewed in this study; yet this appears to be a promising recovery method. Low-temperature data at the low concentrations (and pressures) required for these systems are not available. The mass transfer problems and sorbent life have not been determined. In addition, there are many other sorbents (e.g., zirconium and the rare-earth metals) that may be more practical than yttrium. These materials probably behave similarly to yttrium.

## 6. REJECTED REMOVAL METHODS

Several proposed removal schemes can be rejected as economically infeasible processes under the ground rules adopted in this study. These schemes are discussed below.

## 6.1 Distillation

Both single-stage flash distillation and multistage fractional distillation have been proposed as means for removing tritium;<sup>24</sup> however, scoping studies have shown that distillation is an expensive process with problems related to difficult materials of construction (for fractional distillation) and condenser design (single-stage flash). Distillation was ruled out in the present study for more fundamental reasons. Although meager vapor-liquid equilibrium data are available at the low concentration of tritium required by the ground rules adopted for this study, the equilibrium relations are assumed to follow Sieverts' law. That is,

$$X_T = K \left( P_{T_2} \right)^{1/2},$$

where  $X_T$  is the mole fraction in the liquid and  $P_{T_2}$  is the partial pressure of tritium in the vapor. This equation can also be written in terms of the mole fraction of tritium in the vapor:

$$X_T = \left( K P_M \right)^{1/2} \left( \frac{P_{T_2}^{1/2}}{P_M^{1/2}} \right)$$

$$\approx K P_M^{1/2} Y_{T_2}^{1/2} \approx K' Y_{T_2}^{1/2},$$

where  $P_M$  is the vapor pressure of the bulk metal, lithium, or potassium. The tritium is distilled out of either lithium or potassium. With the low tritium concentrations of interest,

$$P_{T_2} \ll P_M$$

(at least near the feed point), and the total pressure (or  $P_M$ ) is not affected by changes in  $P_{T_2}$  until the tritium pressure becomes large enough to be significant. Note that an azeotrope occurs when

$$Y_{T_2} = X_{T_2} = \frac{X_T}{2},$$

$$\left( \frac{X_T}{2} \right)^{1/2} = k' = K P_M^{1/2},$$

or

$$X_T = 2K^2 P_M.$$

In the lithium stream the azeotrope occurs at  $X_T = 0.05$  at  $1000^\circ\text{C}$  (see Appendix B); in potassium the azeotrope occurs at  $X_T = 10^{-4}$  at  $1000^\circ\text{C}$  (see Ref. 24). These compositions are high by the standards of the present study; that is, the azeotrope compositions are higher than the operating feed compositions. This means that the overhead will actually be enriched in lithium (or potassium) and binary distillation will never produce a composition greater than the azeotrope. Thus a binary distillation appears to be not only expensive but also infeasible.

[Note that the rejection of distillation as a removal scheme is based upon assumed vapor-liquid equilibria at very low tritium compositions, and assumptions could be invalidated by advances in tritium barrier developments or changes in regulatory rules. Nevertheless, it appears unlikely that future developments will make distillation competitive with other recovery schemes.]

## 6.2 Gas Sparging

The reasons for rejecting distillation (discussed above) as a feasible removal system are also used to reject gas sparging. At the compositions of interest to this study, the gas phase would become enriched in lithium (or potassium) rather than in  $T_2$ . Again, the same assumptions that ruled out distillation were used; and, if these are proved incorrect either by experiment or by an advance in tritium barrier

development, the conclusions could be reversed. However, in this case, one could also reject gas sparging without assuming that Sieverts' law holds at these low compositions. The pressures of  $T_2$  over the metals are very low, and the gas sparge rate would be excessive. With a  $T_2$  pressure of  $1.5 \times 10^{-11}$  atm in the lithium system,  $10^{10}$  ft<sup>3</sup> of gas per second would be required to remove 1 kg of  $T_2$  per day. This is an impractical situation, even if the gas phase contained no lithium. Once the tritium is in the gas phase, it is present at such a low concentration that recovery would be expensive. The very high gas rates would require a large sparge vessel and large recovery equipment.

#### 7. RECOVERY OF TRITIUM FROM SYSTEMS WITH HELIUM COOLANT

The potassium coolant and potassium turbine offer several attractive features such as greater thermal efficiency for the system and easier heat removal from the lithium blanket. However, some investigators believe that a helium coolant would be closer to existing technology and, thus, is more clearly feasible at the present time. Fraas<sup>5</sup> has reviewed and compared the advantages and potential problems of both potassium and helium coolants. Use of helium coolant reportedly makes tritium recovery less difficult; yet, the earlier sections of this report have shown that recovery of tritium, even with a potassium coolant, appears to be feasible at reasonable cost.

The detailed proposed designs for these systems are still being developed. The helium can be used simply as a coolant to transfer heat to a steam system, but it could also be used to drive a gas turbine directly. The use of oxygen (trace concentrations) in the helium would probably reduce the tritium pressure to a level where there would be only minor losses of helium to the steam (if a steam turbine is used). However, since the lithium-helium heat exchanger will probably be made of niobium, it would need to be coated with palladium or possibly some other metal on the side that contacts helium if a significant oxygen pressure were used. Corrosion of niobium may be a problem even when only trace concentrations of oxygen are present, e.g., when no oxygen is intentionally added.

If a gas turbine is chosen, the loss to the water coolant system would be even less since the helium-water heat exchanger would be at a very low temperature (100 to 236°F), where the permeability of even stainless steel is low. In a gas turbine system, tritium would be lost principally through bearings and other leaks in the high-pressure gas system. A typical loss rate could be 0.01% or less of the helium inventory per day.<sup>25</sup> This estimate is based upon design specifications for the early Dragon fission reactor system using a helium coolant. However, leakage from the high-pressure gas turbine system need not contaminate a local waterway. The entire turbine should be at least doubly contained; that is, the turbine system should be enclosed in sealed rooms, and the atmosphere in the room should be continuously processed through a dryer to remove tritiated water that may have leaked from the turbine system. A similar arrangement should be used around all components of the blanket and power system because  $T_2$  (if not  $T_2O$ ) will leak or permeate from all surfaces.

If helium is used as a coolant coupled to a steam power generation system, a sorber system could be used just as was suggested with a potassium coolant. If corrosion problems associated with helium coolants containing oxygen are solved, one could remove the tritium as  $T_2O$ . This would lower the tritium pressure (and thus the release rate) drastically. The problems are similar to those discussed in Sect. 3. The niobium surface separating the lithium blanket and the helium coolant would have to be coated on the helium side to prevent rapid oxidation of niobium.

## 8. CONCLUSIONS

The most significant conclusion from this study is that only very low tritium concentrations and pressures can be tolerated in the blanket system presently proposed for fusion reactors. Under the ground rules adopted for this study, the maximum tritium pressure within the blanket system will be approximately  $1.6 \times 10^{-11}$  atm. At pressures in this range, only two of the currently proposed removal methods, tritium diffusion through a metal window and cold traps filled with sorbents, appear to be clearly feasible. Cold traps alone will probably not be economical

since both equilibrium and mass transfer requirements (vessel size) are marginal or unknown. However, by packing the vessel with a sorbent, at least the equilibrium limitations can be removed, and it appears likely that mass transfer rates will be improved. Both processes, however, involve uncertainties, and considerable experimental data are needed to establish their feasibility and to obtain the necessary development and design data.

Considerably lower tritium release rates would be possible by using a "trapping loop." This loop could contain an oxidant to convert the tritium to a nonvolatile form,  $T_2O$ . The loop could contain a  $He-O_2$  mixture or possibly a mixture of nitrate and nitrite salts. As mentioned earlier, a helium coolant itself could constitute the trap loop, but corrosion problems would have to be solved. A separate loop could be included between the coolant and the steam system. Although such a system would reduce tritium losses to the steam system, it would be expensive and would probably not be warranted unless much more severe regulations on tritium release rates are adopted.

#### 9. RECOMMENDED EXPERIMENTAL PROGRAM

Although the results of this study indicate that the recovery of tritium will be feasible at reasonable cost and without prohibitive loss to the environment, considerable chemical and engineering information is needed, and an experimental program is recommended to obtain these data and to check and quantify many of the assumptions made. Such data would allow firm designs for a demonstration system (or systems) to eventually be made.

The experimental program should be directed at the two recovery processes which appear to be economically and technically feasible. Specific engineering studies needed for metal window development are: (1) to demonstrate that the tritium pressure outside the window can be reduced to a value below  $10^{-11}$  atm, (2) to evaluate the film mass transfer resistance in lithium metal in order to confirm the assumed values,

and (3) to investigate materials for the window. Chemical studies are needed to obtain vapor-liquid data for the Li-T<sub>2</sub> system at low tritium concentrations (in the Sieverts' law region) and at higher temperatures (up to 1000°C).

The only other recovery method which warrants experimental work at this time is cold trapping (especially with solid sorbents). Chemical data are needed to determine K-T and K-T-Li solid-liquid equilibria in low-temperature regions (150-250°C). Engineering data are needed to determine mass transfer rates or minimum residence times. If the rates are not considerably higher than those observed in oxide cold traps or if improved cold traps designs are not developed, cold trapping without solid sorbents will probably not be economical. Testing of other solid sorbents for tritium removal in molten salt reactors is currently under way in the Reactor Chemistry Division. These efforts may produce materials or data that will be useful in fusion systems. It is important to make measurements which will, at least by extrapolation, cover the concentration range of interest in fusion systems. Since most metals will probably obey Sieverts' law at low concentrations, extrapolation may be relatively accurate and reliable. All new proposed sorbents should be examined in the same manner described in Sect. 5 (i.e., the maximum loading and trap volume should be evaluated). Engineering studies should be made to evaluate mass transfer and pressure drop in promising systems. These may be the dominant problems in some sorber systems, especially if solid-phase diffusion controls the loading rate.

None of the other proposed removal methods appears to merit further study. Unless the assumptions used in this study are in error, neither distillation nor gas sparging is likely to become an economical process.

The other area suitable for study or deliberation is improvement in the tritium barrier between the steam system and the heat transfer fluid (potassium or helium). Various coating materials are under study in the Molten Salt Reactor Program, and any successful barriers developed in that program can probably be applied to fusion systems. If regulations that limit the tritium release rate to levels substantially less than

4 Ci/day are developed, consideration should be given to adding a trapping loop (He-O<sub>2</sub> or nitrate-nitrite salts) to reduce tritium leakage to essentially zero. In evaluating the cost of trapping loops, credit can be taken for large fractions of the tritium removal costs since these may be reduced considerably, but the major cost would be in the loop itself.

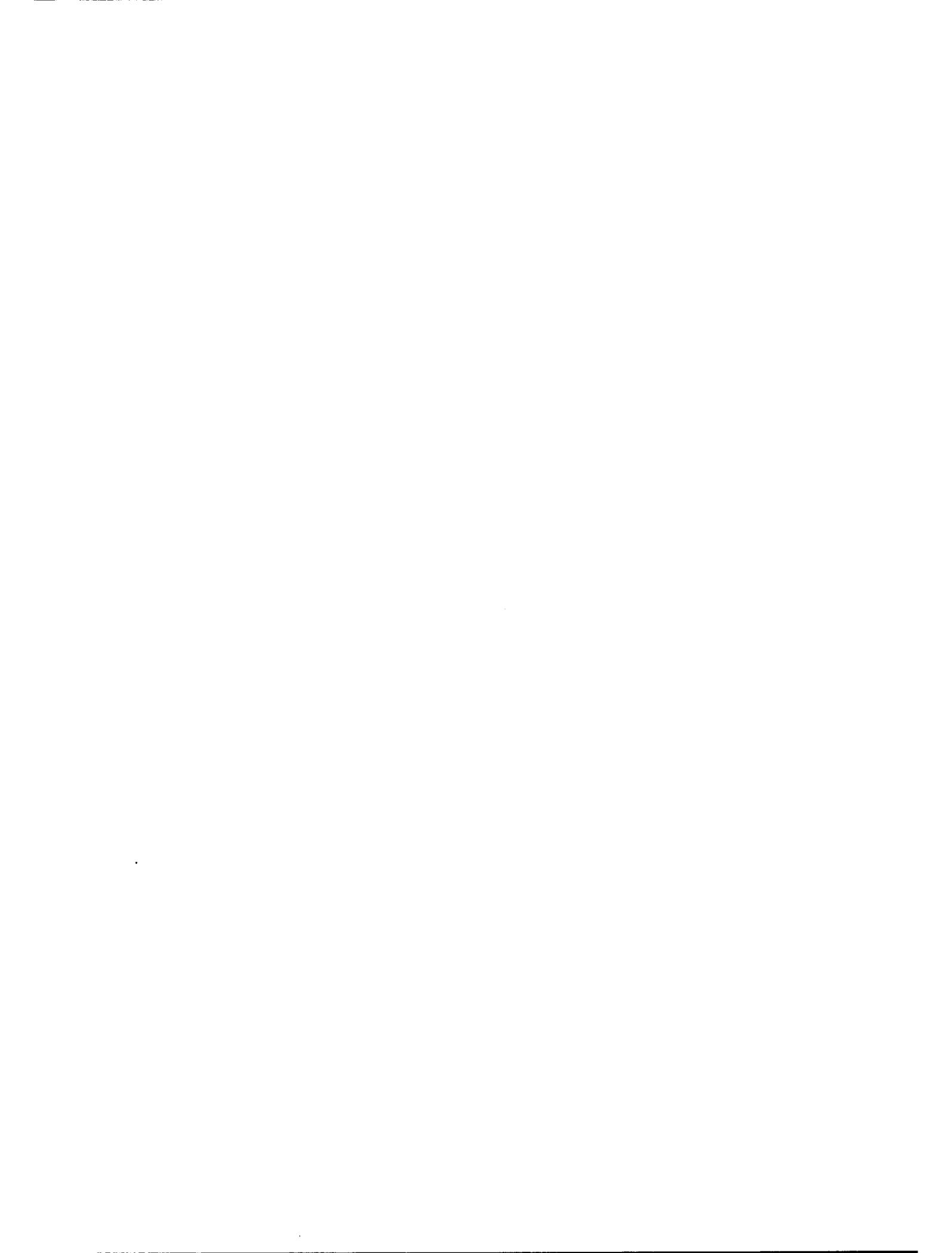
Both the engineering and chemical studies should be coordinated with other programs related to the blanket and power system design. Obviously, a development program for the tritium recovery system should be redirected whenever necessary to support the particular power system that currently appears most promising. It may be less obvious, however, that tritium recovery and containment problems may influence the choice between the more promising power systems. Close association of these efforts with each other, with a plasma program, and with metallurgy or materials support will greatly enhance progress in all phases of the overall effort.

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11. APPENDIXES

### 11.1 Appendix A: Mass Transfer Resistance on Metal Windows

#### 11.1.1 Lithium Flux

Assume the window area is approximately  $1.5 \times 10^7 \text{ cm}^2$ . This estimate came from calculations that assumed no film resistance. Then the flux through the window is:

$$\phi = \frac{\frac{10^3}{6} \text{ g-moles/day}}{8.64 \times 10^4 \frac{\text{sec}}{\text{day}}} \bigg/ 1.5 \times 10^7 \text{ cm}^2$$

$$= 1.29 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}.$$

#### 11.1.2 Mass Transfer Coefficient for Lithium

Assume the diffusion coefficient of tritium in molten lithium at  $1000^\circ\text{C}$  is  $4 \times 10^{-5} \text{ cm}^2/\text{sec}$ .<sup>8</sup> The kinematic viscosity is estimated by extrapolating lower-temperature data (L. M. Handbook) to be  $0.00345 \text{ cm}^2/\text{sec}$ , giving a Schmidt number of 86.3. With a 1-in.-ID channel and a velocity of 2 fps, the Reynolds number is approximately 50,000. The mass transfer coefficient is:

$$N_{\text{Nu}} = \frac{kd}{D} = 0.023 N_{\text{Re}}^{0.8} N_{\text{Sc}}^{1/3}$$

$$N_{\text{Nu}} = 584$$

$$k = 0.0092 \text{ cm/sec}$$

#### 11.1.3 Concentration at the Window Wall

The concentration drop across the liquid metal film would be:

$$\Delta C_{\text{film}} = \frac{1.29 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}}{0.0092 \frac{\text{cm}}{\text{sec}}}$$

$$= 1.4 \times 10^{-8} \frac{\text{g-moles}}{\text{cm}^3}.$$

The bulk concentration at  $1.6 \times 10^{-11}$  atm can be estimated from Ref. 11. If  $k = 0.0078 \text{ mm}^{-1/2}$ , this corresponds to a concentration of  $8.6 \times 10^{-7}$  mole fraction or  $2.8 \times 10^{-7} \text{ g-moles/cm}^3$ . (Note that this concentration is considerably greater than the concentration drop across the film; hence film resistance, although detectable, is not controlling the transfer rate.)

#### 11.1.4 Potassium Liquid

Assume that the window area is approximately  $3.38 \times 10^6 \text{ cm}^2$ . This area is larger than that estimated for lithium because the temperature in the potassium liquid region is less ( $600^\circ\text{C}$  instead of  $1000^\circ\text{C}$ ) and the permeability of palladium is less. The flux through the window is:

$$\phi = \frac{\frac{10^3}{6}}{(8.64 \times 10^4) (3.38 \times 10^6)} = 0.574 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}.$$

#### 11.1.5 Mass Transfer Coefficient for Potassium

Assume again that the diffusion coefficient of tritium in potassium at  $600^\circ\text{C}$  is  $4 \times 10^{-5} \text{ cm}^2/\text{sec}$ .<sup>9</sup> The kinematic viscosity is estimated by extrapolation<sup>26</sup> to be  $0.00214 \text{ cm}^2/\text{sec}$ , giving a Schmidt number of 53.6. The mass transfer coefficient is estimated as follows:

$$N_{\text{Nu}} = 0.023 N_{\text{Re}}^{0.8} N_{\text{Sc}}^{1/3}.$$

If the Reynolds number is 50,000 in a 1-in. channel,  $N_{\text{Nu}} = 498$  and  $k = 0.00785 \text{ cm/sec}$ .

#### 11.1.6 Concentration at the Window Wall

The concentration drop across the liquid film is:

$$\Delta C_{\text{film}} = \frac{0.573 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}}{0.00785 \frac{\text{cm}}{\text{sec}}} = 7.3 \times 10^{-9} \frac{\text{g-moles}}{\text{cm}^3}.$$

The bulk concentration in the potassium stream was estimated in Sect. 2 of this report (for a pressure of  $1.45 \times 10^{-11}$  atm) to be 0.003 wt ppm. This corresponds to a concentration of approximately  $1.4 \times 10^{-9}$  g-atm/cm<sup>2</sup>, which is a small fraction of the estimated concentration drop across the liquid film. This indicates that film resistance will be the principal resistance affecting the transfer rate. By adding approximately 4 at. % lithium, Fraas suggests that the concentration of tritium could be varied by a factor of  $\sqrt{10}$ . However, even this improvement would not make the potassium stream the best place to locate the metal window.

#### 11.1.7 Potassium Vapor

If the permeability of palladium controls the transfer rate through the metal window, the flux will be the same as in the lithium system because the temperature is essentially the same. (However, the higher pressure in the potassium vapor region could require thicker tubing and thus lower fluxes.) For example,

$$\phi \approx 1.29 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}.$$

#### 11.1.8 Mass Transfer Coefficient in Potassium Vapor

Assume that  $\mu = 0.053$  lb/ft-hr or  $0.000219$  g/cm-sec (Ref. 18, p. 30), and the density of the vapor is  $5.2 \times 10^{-5}$  g/cm<sup>3</sup>. Then the kinematic viscosity is  $0.108$  cm<sup>2</sup>/sec.

The diffusion coefficient can be estimated by the method proposed by Gilliland and cited by Perry.<sup>13</sup>

$$D_G = 0.0043 \frac{T^{3/2}}{P (V_1^{1/3} + V_2^{1/3})^2} \sqrt{\frac{1}{M_1} + \frac{1}{M_2}}.$$

The values and units used in this expression to estimate  $D_G$  are given below:

$$T = 1273^\circ\text{K}$$

$$P = 4140 \text{ mm (5.447 atm)}$$

$$V_1 = 7.15 \text{ cc/g-mole (assumed to be the same as hydrogen)}$$

$$V_2 = 39/0.67 \text{ cc/g-atom (assumed to be the same as liquid potassium)}$$

$$M_1 = 6$$

$$M_2 = 39$$

Then  $D_G = 0.467 \text{ cm}^2/\text{sec}$ . The Schmidt number is 0.232.

Assume that the Reynolds number is 10,000 in 1-in. tubes. This corresponds to a velocity of 90 cm/sec. Then

$$\begin{aligned} N_{\text{Nu}} &= 0.023 (10,000)^{0.8} (0.232)^{1/3} \\ &= 22.4, \end{aligned}$$

or  $k = 4.115 \text{ cm/sec}$ .

#### 11.1.9 Concentration at the Window Wall (in Potassium Vapor)

The concentration drop across the gas film is:

$$\Delta C_{\text{film}} = \frac{1.29 \times 10^{-10} \frac{\text{g-moles}}{\text{sec-cm}^2}}{4.115 \frac{\text{cm}}{\text{sec}}} = 3.13 \times 10^{-11} \frac{\text{g-moles}}{\text{cm}^3}.$$

To estimate the tritium concentration in the potassium vapor, assume that the vapor is an ideal gas with a tritium pressure of

$$\begin{aligned} (1.45 \times 10^{-11} \text{ atm}) \frac{5.447 \text{ atm} - \text{vapor pressure}}{115/760 \text{ atm} - \text{condenser pressure}} \\ = 5.22 \times 10^{-10} \text{ atm.} \end{aligned}$$

$$pv = RT = (82.06 \frac{\text{cc-atm}}{\text{°K-mole}}) (1273^\circ\text{K}),$$

and  $p = 5.22 \times 10^{-10} \text{ atm};$

thus  $C_{T_2} = \frac{1}{v} = 4.98 \times 10^{-15} \frac{\text{g-moles}}{\text{cm}^3}.$

Therefore, the mass transfer rate would be completely controlled by the vapor film resistance, and the metal window should not be located between the Li-K heat exchanger and the water boiler, where the potassium is in the vapor state.

### 11.2 Appendix B: Estimation of Azeotrope Composition in Lithium

As discussed in Sect. 6.1, the azeotrope occurs when

$$X_T = 2 k^2 P_M,$$

$$k = \frac{X_T}{P_T^{1/2}}.$$

Since there are no data on the lithium-tritium vapor-liquid equilibrium above 825°C and at low compositions, the existing data must be extrapolated to 1000°C. Assume that

$$K \propto \exp (1/T).$$

The low-pressure data of Messer<sup>11</sup> were thus extrapolated to 1000°C. The data are scattered (they come from several experimenters); however, one can estimate a value for K of approximately  $0.0078 \text{ mm}^{-1/2}$  at 1000°C. The uncertainty in this value appears to be approximately  $\pm 50\%$ , but any value within this range would lead to the conclusions reported in Sect. 6.

Although not mentioned in this report, the azeotrope in the potassium-tritium system can be seen from Farookhi and Rogers.<sup>24</sup> The azeotrope was estimated to occur at a tritium atom fraction of approximately  $10^{-4}$ .

### 11.3 Appendix C: Extrapolation of Yttrium-Hydrogen (Tritium) Temperature - Pressure Data

The lowest composition data points for the temperatures shown in the insert of Ref. 23 are as follows:

x (at. %)	$\sqrt{p_{T_2}}$ (mm <sup>1/2</sup> )	$\left(\frac{\sqrt{p_{T_2}}}{\text{at. \%}}\right)$	T (°C)	°K <sup>-1</sup> x 10 <sup>3</sup>
9.5	0.23	2.42	949.4	0.818
6.7	0.10	1.49	899.3	0.853
6.3	0.06	0.952	849.9	0.891
10.0	0.065	0.65	799.9	0.932
6.9	0.035	0.507	750.3	0.977
7.5	0.015	0.20	700.7	.027
7.8	0.01	0.133	651.1	1.082

The results at low temperatures shown in Ref. 23 are not accurate since it was difficult to read the figure of Yannopoulos et al. These data are plotted in Fig. C-1. As expected, the data can be fit reasonably well by a straight line.

$$\ln_e \frac{p_{T_2}}{X_T} = 9.956 - \frac{11.12 \times 10^3}{T}$$

If  $p_{T_2}$  is taken to be  $1.45 \times 10^{-11}$  atm at 600°C, it will be less at T. Compere et al.<sup>10</sup> report, for potassium:

$$\log_{10} p = 11.69 - \frac{6175}{T},$$

$$\log_{10} \frac{p_{T_2}}{p_{823}} = 7.073 - \frac{6175}{T},$$

$$\ln_e \frac{p_{T_2}(T)}{p_{873}} = 16.287 - \frac{14,218}{T},$$

$$\ln_e p_{T_2}(T) = -8.670 - \frac{14,218}{T},$$

where  $p_{T_2}$  is given in mm Hg. Then

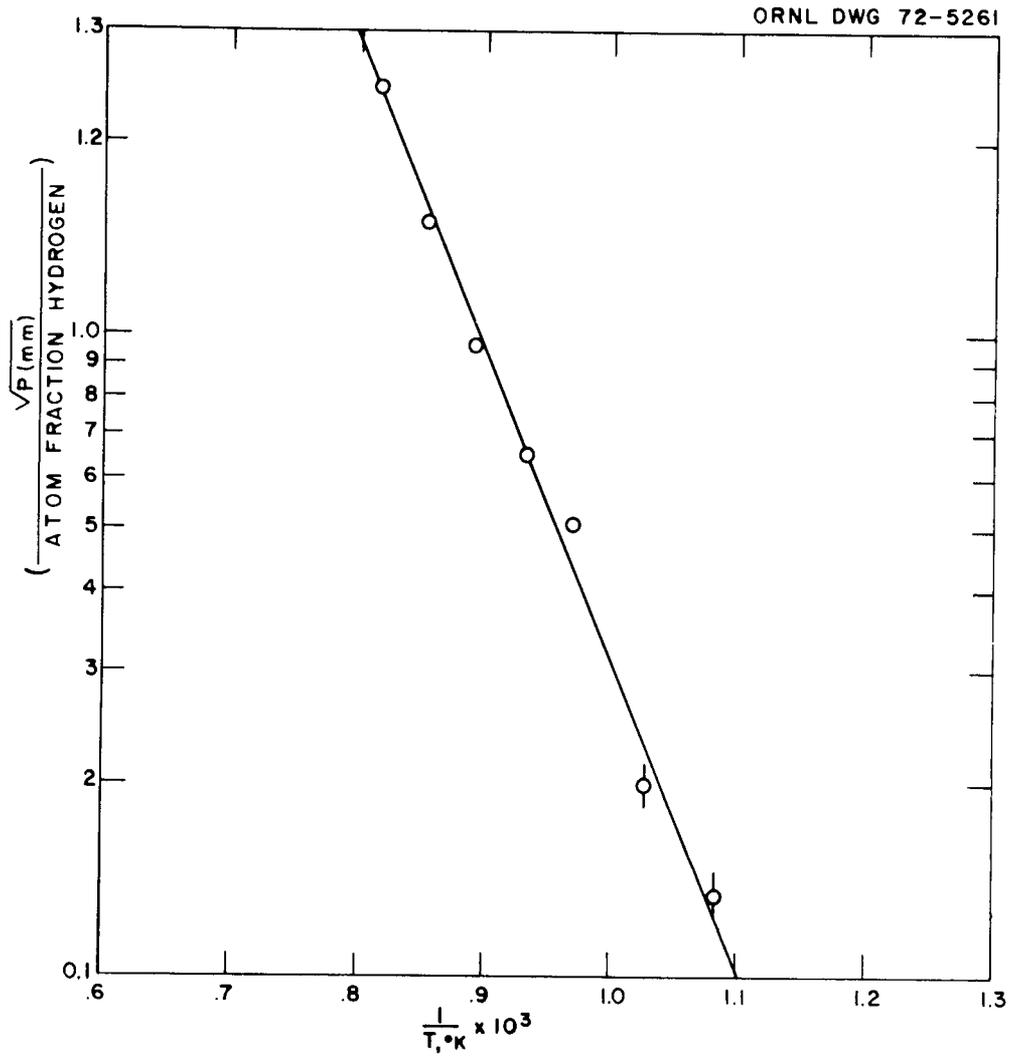


Fig. C-1. Hydrogen Sorption on Yttrium (Data from Yannopoulos et al.).

$$\begin{aligned} \ln_e (X_T) &= 1/2 \ln_e (p_{T_2}) - 9.956 + \frac{11,120}{T} \\ &= -14.291 + \frac{4011}{T} ; \end{aligned}$$

at 82°C,  $X_T \approx 0.05$ .

#### 11.4 Appendix D: Tritium Leakage Through the Regenerator Walls

Neglect liquid film resistance and assume that the permeation constant of the metal wall changes with temperature in the following manner:

$$K_{ss} = A_1 \exp (-A_2/T).$$

Then the concentration in the metal wall is:

$$C = k \sqrt{p}.$$

Assume for  $k$ , the value corresponding to 600°C. Although  $k$  will increase with decreasing temperature, this effect will be ignored. Thus a conservative answer will be obtained (i.e., the tritium leakage rate will be overestimated).

A material balance shows that

$$\begin{aligned} -\frac{dC_1}{dz} &= \frac{\phi A}{v} = \frac{AK_{ss} (\sqrt{p_1} - \sqrt{p_2})}{vt} \\ &= \frac{K_{ss} A}{tkv} (C_1 - C_2) \\ &= \frac{AA_1 \exp (-A_2/T)}{kvt} (C_1 - C_2). \end{aligned}$$

If the cross-sectional areas for flow on both sides of the exchanger are equal,

$$C_1 - C_2 = \text{constant} = \Delta C.$$

Assume that metal entering the regenerator from the water boiler (e.g., at the hot end of the regenerator or at  $Z = 0$ ) will be at temperature  $T_0$  and the metal leaving the regenerator to enter the secondary exchanger and cold trap (at  $Z = 1$ ) will be at temperature  $T_1$ . Since the temperature varies linearly along the regenerator path, the temperature at any point can be expressed as follows:

$$T = T_0 - \frac{T_0 - T_1}{\ell} Z.$$

The material balances can be written as

$$\int_{C_0}^{C_1} dC = - \frac{AA_1 \Delta C}{kvt} \int_0^1 \exp - \left( \frac{A_2}{T_0 - \frac{T_0 - T_1}{\ell} Z} \right) dZ$$

or

$$\int_{C_0}^{C_1} dC = \frac{AA_1 \Delta C}{kvt (T_0^* - T_1^*)} T_0^* \int_{T_0^*}^{T_1^*} \exp \left( \frac{1}{T^*} \right) dT^*,$$

where  $T^* = T/A_2$ . The integral on the right-hand side was approximated graphically over the short range needed. The following values were estimated for quantities maintained above:

$$v = 60.96$$

$$t = 0.3 \text{ min}$$

$$A = 1.57 \text{ cm}^2/\text{cm}^3 \text{ (for 1-in.-OD)}$$

$$\ell = 10^3 \text{ cm}$$

$$\begin{aligned} A_1 &= 1.5 \times 10^{-3} \text{ torr} \cdot \text{liters/cm} \cdot \text{sec} \cdot \text{torr}^{1/2} \\ &= 8.81 \times 10^{-8} \text{ g-mole/cm} \cdot \text{sec} \cdot \text{torr}^{1/2} \end{aligned}$$

$$A_2 = 1582^\circ\text{K}$$

$$k = 8.73 \text{ ppm/torr}^{1/2} = 2.08 \times 10^{-6} \frac{\text{g-moles/cm}^3}{\text{torr}^{1/2}}$$

$$T_0 = 873^\circ\text{K}$$

$$T_1 = 355^\circ\text{K}.$$

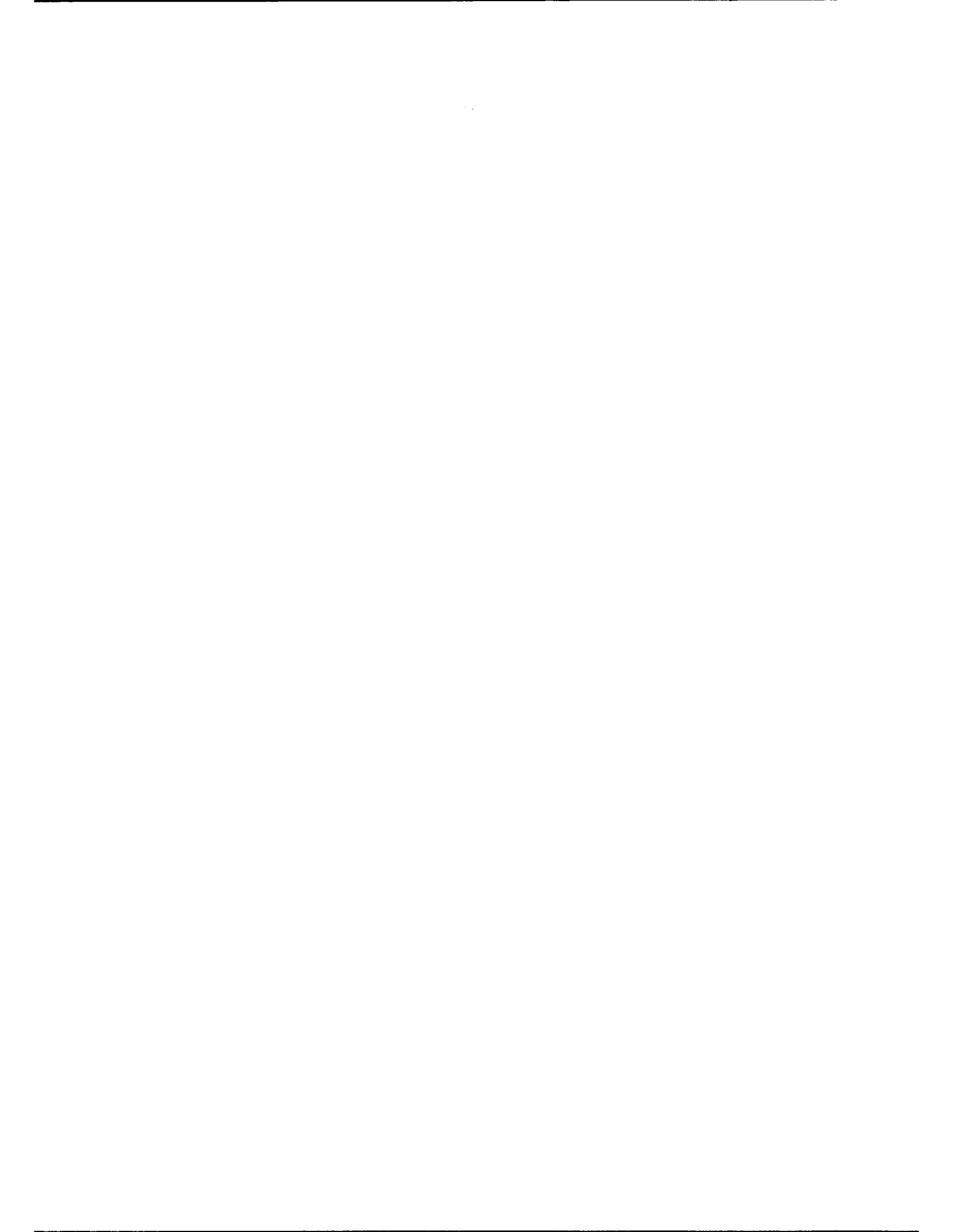
Then

$$\frac{\Delta C}{C_1 - C_0} = \frac{kv(T_0^* - T_1^*)}{AA_1 I},$$

where  $I = \int_{T_0^*}^{T_1^*} \exp -\frac{1}{T^*} dT^* = 4 \times 10^{-11},$

and  $\frac{\Delta C}{C_1 - C_0} = 10^{16}.$

Thus there is very little change in concentration along the flow path until the potassium actually enters the cold trap. If we had taken into account the change in tritium pressure with temperature, the loss would have been even less.



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