

TRITIUM PROBLEMS IN FUSION REACTOR SYSTEMS*

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ABSTRACT

A brief introduction is given to the role tritium will play in the development of fusion power. The biological and worldwide environmental behavior of tritium is reviewed. The tritium problems expected in fusion power reactors are outlined. A few thoughts on tritium permeation and recent results for tritium cleanup and CT_4 accumulation are presented. Problems involving the recovery of tritium from the breeding blanket in fusion power reactors are also considered, including the possible effect of impurities in lithium blankets and the use of lithium as a regenerable getter pump.

INTRODUCTION

Rather than report current results, my purpose in this talk is to emphasize the great diversity of problems before us. I will mention several topics that I have discussed in previous meetings and publications^{1,2} and briefly review a few current research projects.

Tritium-related problems may be divided into three broad categories: containment, recovery, and recycling. Containment includes all the steps we must take to minimize the release of tritium into the environment. To maintain fusion power's current favor among environmentalists, we will have to work very

hard in this area. Major sub-categories of tritium containment include permeation and gas-cleanup processing. Recovery encompasses a broad range of materials problems that must be solved before we can confidently design processes to extract bred tritium from the lithium-containing breeding blanket. These processes must strike some compromise between our minimum tritium-inventory desires and a reasonable cost for processing equipment. Recycling involves the processes required to move the fuel within a fusion reactor, recognizing that much of the fuel will not burn before it is removed from the reactor. The fuel must be pumped, injected, purified, stored, and isotopically separated.

CONTAINMENT

In Figure 1, a cartoon is shown that was first presented by David Rose at the Oak Ridge National Laboratory meeting. I hope that fusion power has not been oversold in this respect; although it will be hard to live up to this vision, we should certainly try. In Figure 2, worldwide tritium inventory through the turn of the century is projected. The bulk of today's tritium is a residual from atmospheric testing of nuclear weapons; natural inventory amounts to only several kilograms and the

*Work performed under the auspices of the U. S. Atomic Energy Commission.

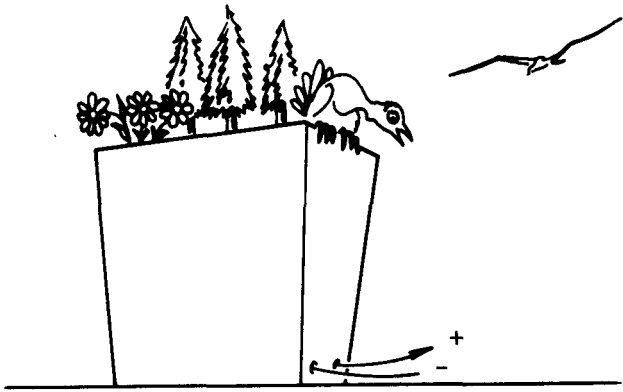


FIGURE 1 - Environmentalist's view of the fusion reactor.

accumulation from fission power is increasing rapidly. There is no expected contribution from fusion power in this time frame. Based on these inventory projections, individual dose estimates have been made in the neighborhood of 0.03 mrem/yr, a trivial amount.³

Of greater concern is the dose people might receive within or in the immediate vicinity of a large tritium-containing facility.

Recently, I had the opportunity to participate in a major design study for a fusion engineering research facility (FERF). With once-through-ventilation, it was postulated that a credible accidental release was 10 g of tritium in the chemical form HTO in a 600-sec time span. The dose people could receive by inhalation and skin absorption is shown in Figure 3. These predictions were based on numerical models that now form the basis of the Atmospheric Release Advisory Capability (ARAC) maintained by the AEC at Livermore. You may have seen a reference to this in the September issue of Industrial Research, where mention was made of our modeling the accidental release at the Savannah River Plant last May 2. From the predicted dose contours, one clearly

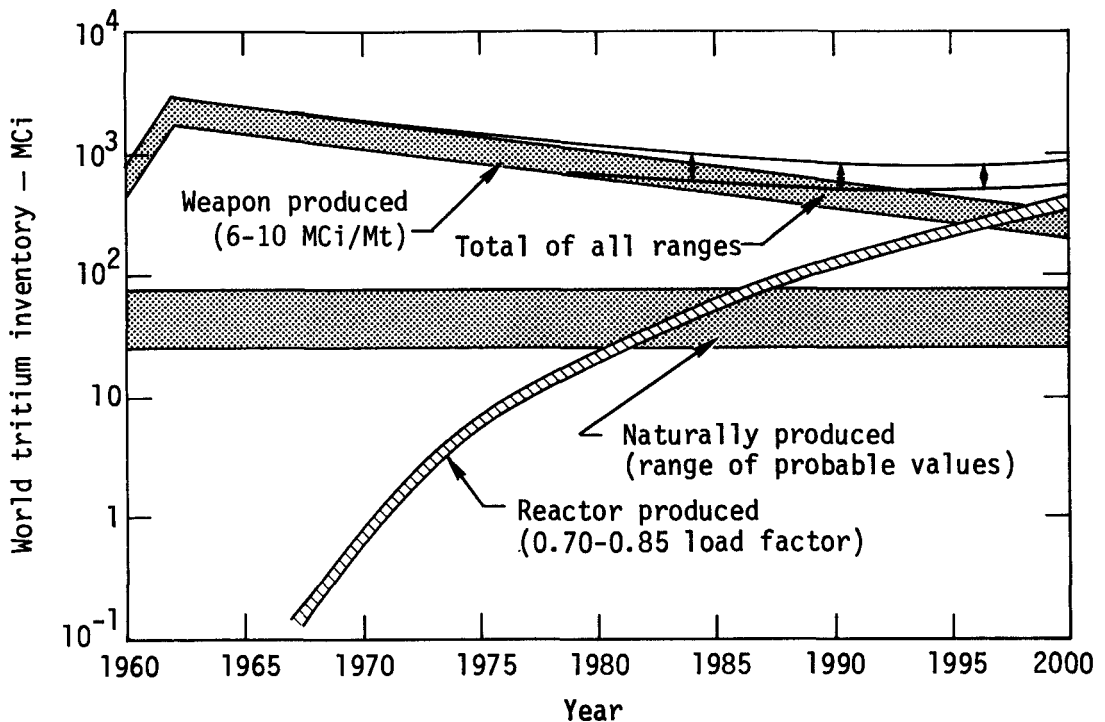


FIGURE 2 - Projected worldwide tritium inventory contribution through the year 2000.

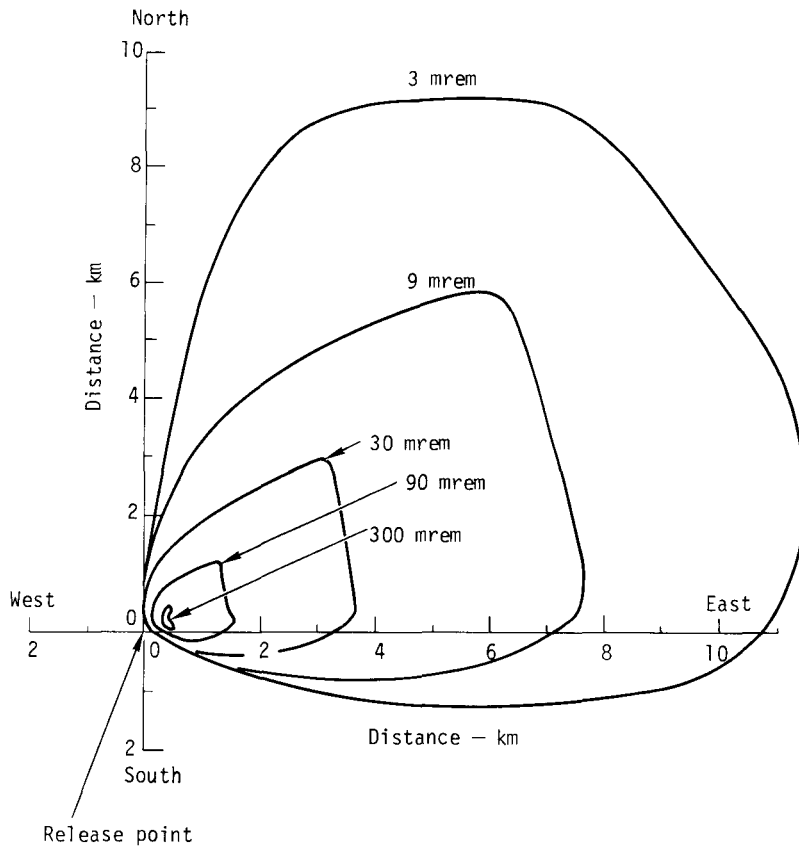


FIGURE 3 - Estimated radiation dose for summer weather at LLL by inhalation and skin absorption from 10 g of tritium released as HTO.

could receive a dose in excess of 5 mrem at distances as far away as a few kilometers. Therefore, the strategy adopted for our FERF design was one of total enclosure. This is the subject of another report.⁴

Another aspect of containment involves permeation of tritium through hot metals, particularly heat-transfer surfaces. The permeabilities of a few materials are shown in Figure 4.¹ Tungsten is generally believed to be the metal with the lowest permeability, and we see that it is about the same as the oxide with one of the highest permeabilities. For materials with comparable chemical affinity (solubility) for tritium, we expect that permeability will be proportional to diffusivity.

In Figure 5, diffusivities of hydrogen in some oxides are shown. On this basis, beryllia is

predicted to be less permeable than alumina by a factor of 10^7 . Imposing an oxide permeation barrier in heat-transfer equipment would be difficult. Making a thin film pore free is a demanding task. In the case of beryllia, however, one could use a thicker layer of oxide with little heat-transfer penalty because of its unusually high thermal conductivity.

As shown in Figure 6, the thermal conductivity of beryllia exceeds that of stainless steel over the temperature range in which they might be used. A technology for such composite materials does not presently exist, but if it did, permeation losses to the environment could be all but eliminated. A substantial technology for fabrication of BeO does exist, however, because several years ago, LLL built the larger part of a nuclear reactor from it.

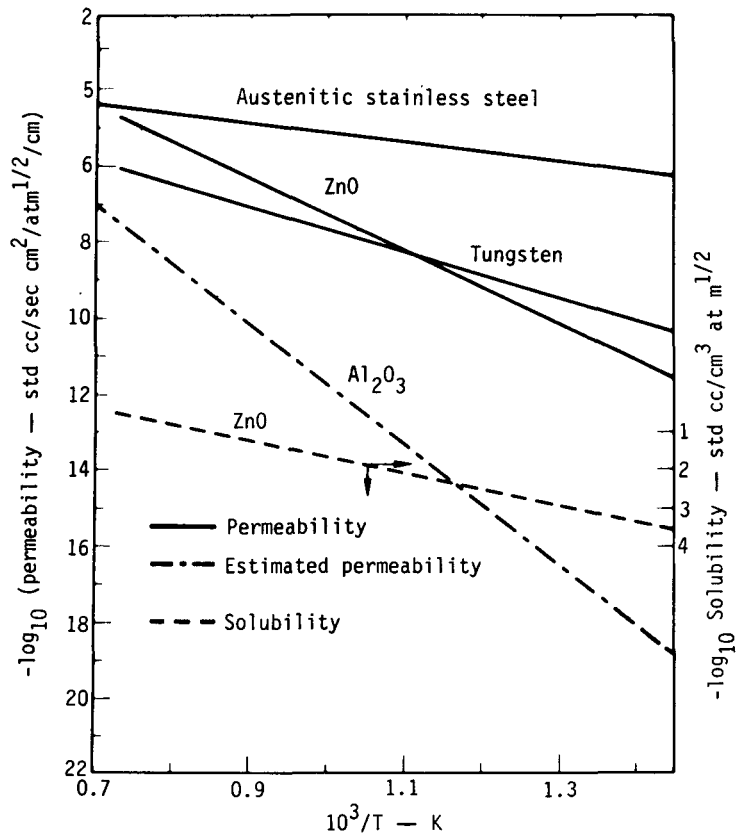


FIGURE 4 - Measured and predicted permeabilities for selected materials.

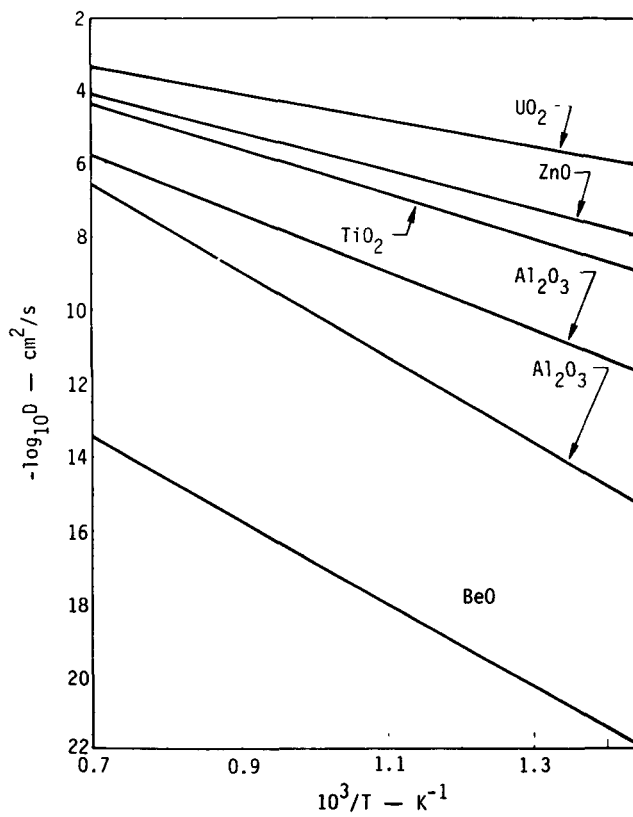


FIGURE 5 - Diffusivities of hydrogen in various oxides.

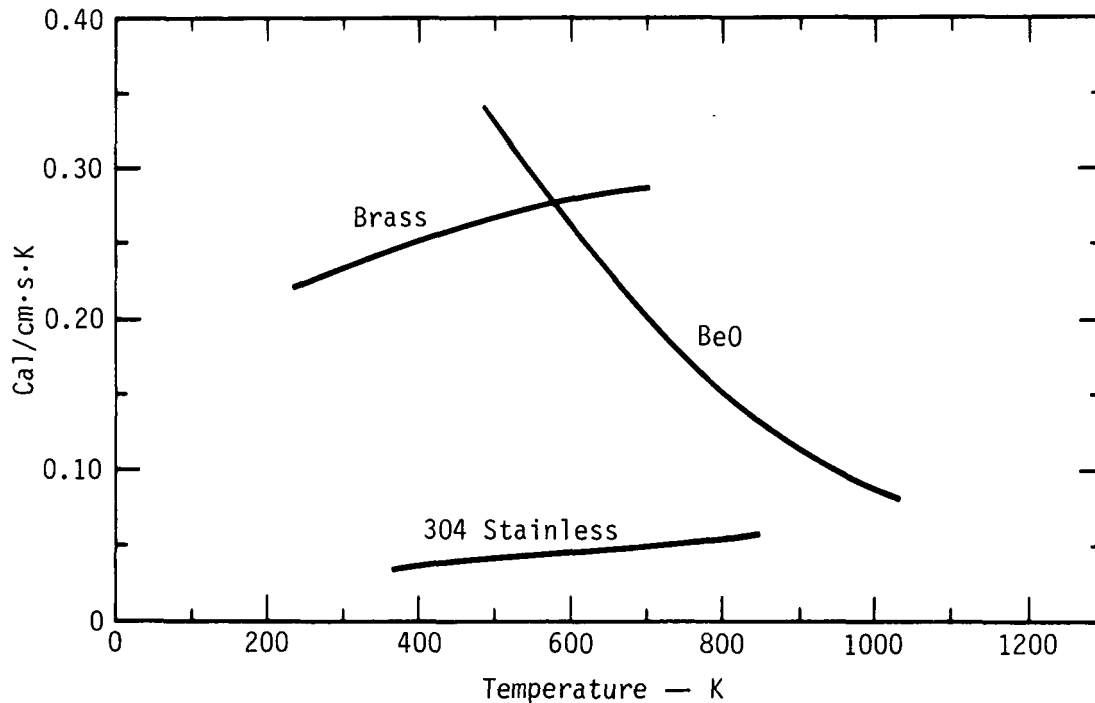


FIGURE 6 - Thermal conductivity of beryllia as compared with stainless steel and brass.

One last topic in the category of containment has to do with the cleanup of an inert gas atmosphere into which a substantial quantity of tritium has been accidentally released. This is a current research project at LLL and is part of a \$2.5 million safety/environmental program to upgrade our equipment. C. L. Folkers heads the project, and provided Figure 7, showing the performance of a bed of activated uranium through which argon containing 1% D₂ is flowed. The quality of cleanup depends on the flow rate and chemical stoichiometry of the bed. Such performance is intuitively reasonable, but is also a dramatic demonstration of the fact that the solid-gas system is not reaching equilibrium. To expect that chemical processes depending on solid-state diffusion can be accurately designed on the assumption of chemical equilibrium is naive. In another experiment, a flow-through uranium bed was

used to strip hydrogen from helium down to hydrogen concentrations below 1 ppm, but the flow rate was very slow.

RECOVERY

Another of David Rose's cartoons, Figure 8, shows what a tritium breeder's parochial point of view might be: fuel, lithium, and magnetic field for input; tritium, lithium, and power for output. Processing details are left to the imagination. Peoples' imaginations have been busy, though, and several conceptual processes have been put forth. They usually depend on chemical gettering, cold trapping, selective permeation, extraction, or some combination of these techniques.

One process that has appealed to me uses a combination of permeation and chemical gettering. The most popular permeation membrane to date

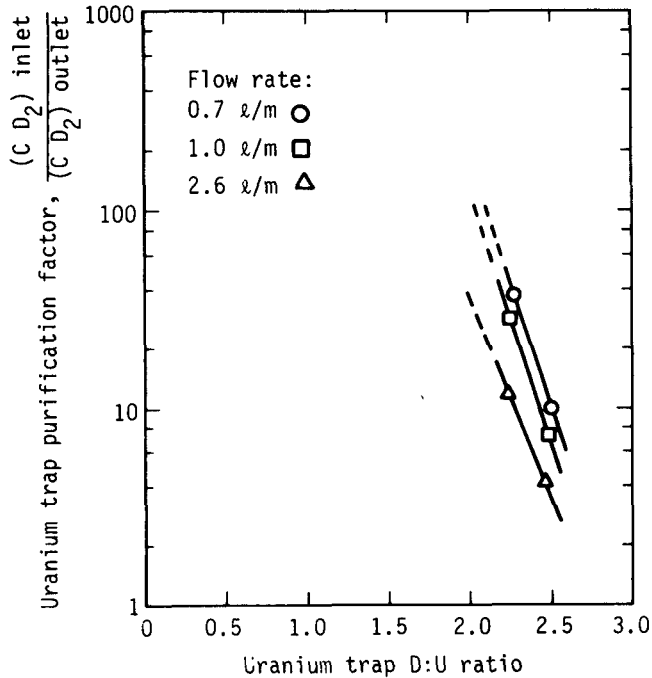


FIGURE 7 - Uranium-trap performance as a function of its D:U ratio. All flow conditions with 1 vol % D_2 in argon.

seems to be niobium, due to its high permeability, good chemical compatibility with lithium, and its high-temperature-operation capability.

In Figure 9, the oft-cited data compiled by Webb is shown.⁵ The right-hand branch of the curve is usually rejected as poor data. The left-hand branch, however, has been used by others in conceptual designs of recovery processes. Note how the permeability appears to increase as the temperature increases. In Figure 10, the solubility data of three authors are nicely represented by a single equation. Diffusivity of hydrogen in niobium shows good agreement between authors who used measurement techniques such as inelastic neutron scattering or the Gorsky effect.⁶⁻⁹ They show a

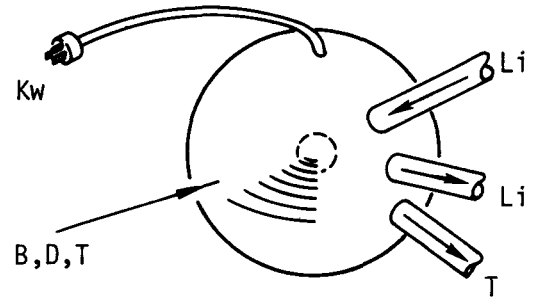


FIGURE 8 - Tritium breeder's view of the fusion reactor.

uniformly low activation energy for diffusion. Using a classical approach, where the hydrogen must pass through the solid-gas interface, a much higher value is observed. If we assume the bulk measurements are more nearly correct and combine them with the solubility behavior as in Figure 11, we find that the permeability, $P = DS$, is predicted to decrease as the temperature increases. The discrepancy between Webb's data and our predictions is still unresolved, although T. Elleman has given us new insights into the classical approach at this conference.

If we continue, confident that predictable permeation will occur, the combination recovery concept can be put forth. It relies on

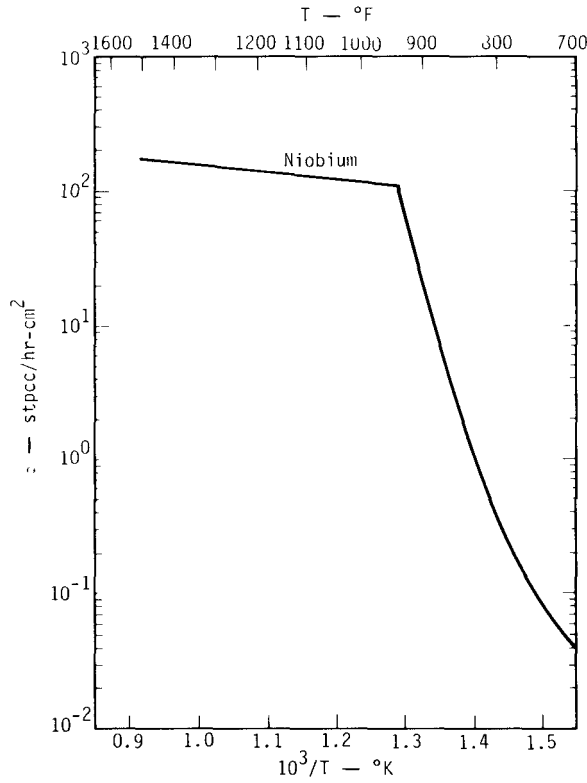


FIGURE 9 - Niobium permeability of hydrogen as a function of temperature.

$$S = 1 \times 10^{-3} \exp\left(\frac{4244.7}{T}\right) \frac{\text{mg}}{\text{cm}^3 \cdot \text{Pa}^{1/2}}$$

$$D = 0.0215 \exp(-9370 \pm 600/RT) \text{ cm}^2/\text{s}$$

$$D = 5.4 \times 10^{-4} \exp\left(\frac{-2510}{RT}\right) \text{ cm}^2/\text{s} \quad (\text{Cantelli})$$

$$D = 5.4 \times 10^{-4} \exp\left(\frac{-2540}{RT}\right) \text{ cm}^2/\text{s} \quad (\text{Schaumann})$$

$$D = 5.0 \times 10^{-4} \exp\left(\frac{-2440}{RT}\right) \text{ cm}^2/\text{s} \quad (\text{Schaumann})$$

$$D = 1.2 \times 10^{-3} \exp\left(\frac{-2710}{RT}\right) \text{ cm}^2/\text{s} \quad (\text{Doremus})$$

FIGURE 10 - Solubility and diffusivity equations of hydrogen in niobium.

$$D = 1 \times 10^{-3} \exp\left(\frac{-3000}{RT}\right) \text{ cm}^2/\text{s}$$

$$P = DS = 1 \times 10^{-3} \exp\left(\frac{-3000}{RT}\right) 1 \times 10^{-3} \exp\left(\frac{4244.7}{RT}\right) \frac{\text{cm}^2}{\text{s}} \cdot \frac{\text{mg}}{\text{cm}^3 \cdot \text{Pa}^{1/2}}$$

$$P = 1 \times 10^{-4} \exp\left(\frac{5404}{RT}\right) \frac{\text{mg}}{\text{Pa}^{1/2} \cdot \text{m} \cdot \text{s}}$$

FIGURE 11 - Combination of solubility and diffusivity of hydrogen in niobium to predict permeability.

the marked temperature coefficient of solubility for hydrogen in molten lithium. By "equilibrating" two flowing lithium streams at different temperatures separated by a permeable membrane and vacuum, we can obtain unusually high enrichments per stage. The chemical route on a PCT diagram and a possible hardware configuration are shown in Figures 12 and 13.²

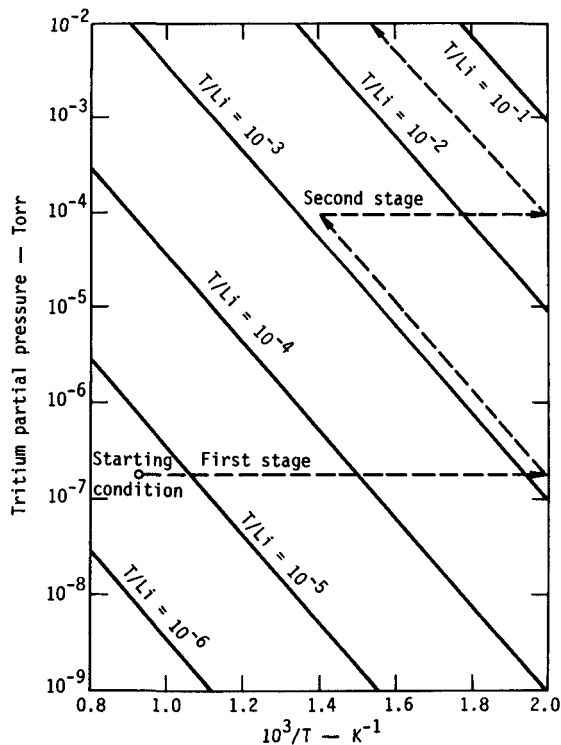


FIGURE 12 - Chemical "route" of a conceptual process for tritium recovery from molten lithium.

Equilibrium will not be achieved, of course, but the figures illustrate the concept.

Another topic that might be worth studying is the effect that dissolved impurities could have on tritium recovery processes.* A few years ago, we had another reactor program at LLL. The reactor was to operate at very high temperature (tungsten-rhenium construction) and was lithium cooled. We had occasion to acquire some high-quality equipment for working with liquid lithium. Subsequently, we did some work on purifying this alkali metal by chemical gettering with yttrium and other metals. The best we ever did was about 15 ppm oxygen.

When oxygen and hydrogen are present in liquid sodium, the chemical activity of each impurity is suppressed by the other because of interaction in the solution. If this same behavior were to be exhibited in the lithium system, the difficulties associated with tritium recovery from the breeding blanket would be compounded.

*Personal communication with V. Maroni since the presentation indicates this should not be a problem.

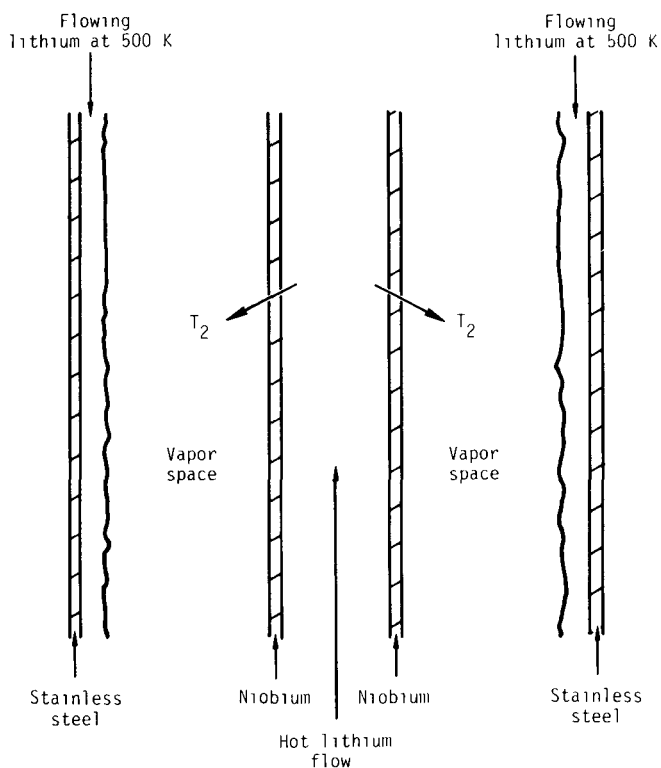


FIGURE 13 - Hardware configuration for one stage of a conceptual process for tritium recovery.

RECYCLING

The problems in recycling operations are fairly well in hand compared with the other two categories. For the most part, they seem to be resolvable by modest extensions of existing technology. There is one problem, however, which we are currently working on that could prove to be a considerable nuisance: the gradual radiolytic production of methane in low-pressure, tritium-containing vessels. This is being studied by V. P. Gede, and his preliminary results are shown in Figure 14 and Table 1.

In a reactor, we expect fairly high surface-to-volume ratios and contact times of months or years during which tritium is present. Accumulation of methanes has been known for many years, but Gede's are the first quantitative results. The rate of accumulation

apparently is dependent on construction materials, surface treatment, and cleanliness. It is likely that temperature and pressure will also be important parameters. Fuel-processing systems will have to be designed to handle this problem, which will probably be a consideration in the first-generation D-T burners. Recovery problems, in contrast, are further in the future.

CONCLUSIONS

These problems in containment, recovery, and recycling are just a sample of the tasks before us. Many more problems exist and will surely come up in the workshops. Despite their number and complexity, however, I am confident they can and will be solved.

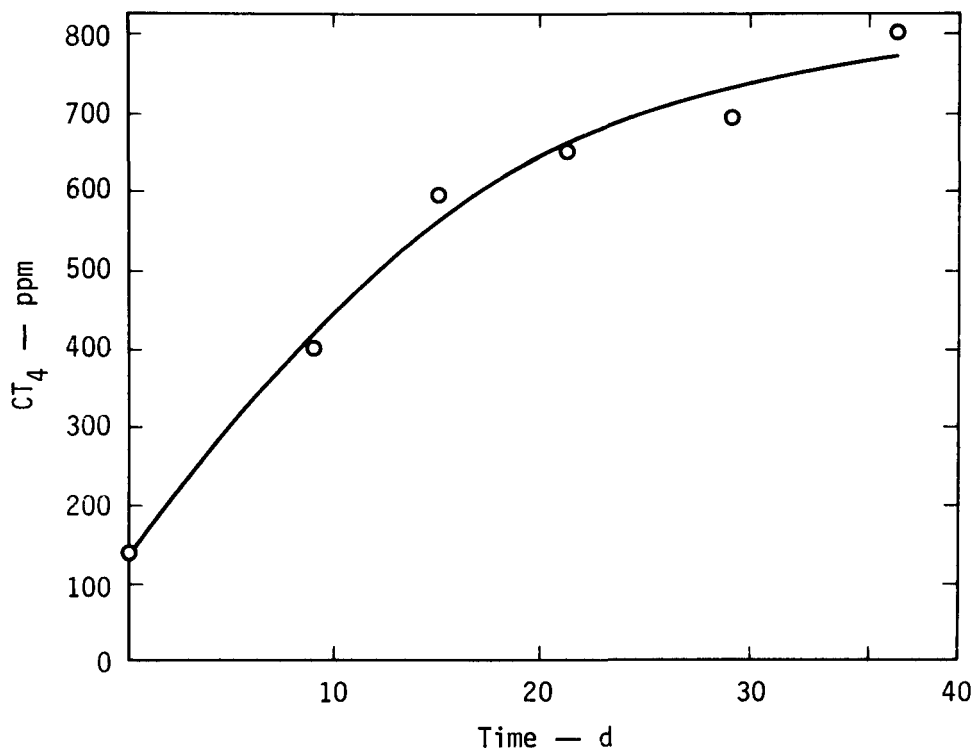


FIGURE 14 - Radiolytic production of methane by tritium as a function of time, using a Western Air Products Co. OFHC copper vessel with Nupro SS-4BK valve (Kel-F seat). Passive, low-memory, sample vessel. (Reference to a company or product does not imply approval or recommendation of the product by the University of California or the U. S. Energy Research and Development Administration to the exclusion of others that may be suitable.)

Table 1

TRITIUM EXPOSURE TESTS [TRITIUM PRESSURE ~100 kPa (1 atm),
TEMPERATURE = 22°C] SHOWING RADIOLYTIC METHANE PRODUCTION

Sample	CH ₄ production
316 SS HOKE sample vessel electropolish	50 ppm ————— 700 ppm ----- 120 d
316 SS HOKE sample vessel, NaOH degrease, glass bead blast 1:1 HNO ₃ + 10 g H ₃ PO ₄ pickle	50 ppm ————— 3600 ppm -----
OFHC copper Western Air Products Co. Low-memory sample vessel with Nupro SS - 4BK valve (Kel-F seat disk)	100 ppm 800 ppm ----- 40 d
304 SS sample vessel with 0.1 g DC-55M grease	20 ppm ————— 42,000 ppm ----- 100 d
304 SS sample vessel control	20 ppm ————— 8300 ppm ----- 100 d

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