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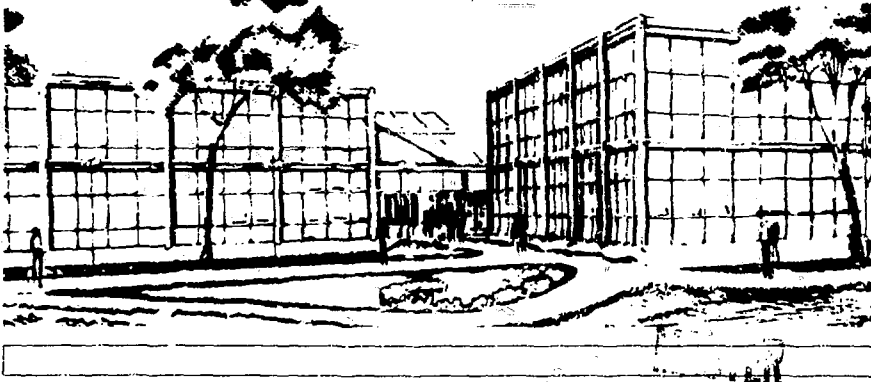
FUEL CYCLE PROBLEMS IN FUSION REACTORS

R. G. Hickman

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# FUEL CYCLE PROBLEMS IN FUSION REACTORS\*

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R. G. Hickman

Lawrence Livermore Laboratory, University California  
Livermore, California 94550

## ABSTRACT

Fuel cycle problems of fusion reactors evolve around the breeding, recovery, containment, and recycling of tritium. These processes are described, and their implications and alternatives are discussed. Technically, fuel cycle problems are solvable; economically, their feasibility is not yet known.

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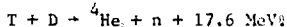
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## INTRODUCTION

Tritium and deuterium are heavy isotopes of hydrogen that can undergo nuclear fusion with relative ease. This process is expressed as:



Tritium is a necessary fuel for thermonuclear reactors and is at the heart of all fuel cycle problems.

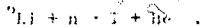
Because tritium has a half-life of only 12.3 y,<sup>1</sup> it is not found naturally in useful concentrations and must, therefore, be bred, usually in nuclear reactors. This is expensive - about \$10,000/g - but new production reactors have not been built for decades.

Tritium is also a hazard to man and the environment. Although the biological effectiveness of its radiation is unity (the same as gamma rays), chronic, low-level exposure to tritium is believed to be three times as radiotoxic as gamma rays for the same absorbed dose.<sup>2</sup> The maximum permissible concentration (mpc) in air is about one part per trillion. A short-term, lethal dose may be received from a single breath of tritiated water vapor or from a single drop of tritiated liquid on the skin. Absorption of tritium through the lungs or skin occurs with equal ease. The biochemical interactions of tritium have been studied extensively<sup>3</sup> and are fairly well understood.

The chemical behavior of tritium is nearly identical to that of normal hydrogen: it burns, explodes, permeates and embrittles metals,<sup>4</sup> and can be incorporated into biological molecules.

## BREEDING TRITIUM

Tritium is usually bred in nuclear reactors using lithium as the breeding material. Lithium can absorb a neutron and produce tritium and helium:



Fusion reaction (1) produces a neutron that can be used to create more tritium. Other breeder reactions include those of  ${}^7\text{Li}$  and the neutron-multiplying reaction of beryllium:



The breeding material is contained in a "blanket" layer just outside the innermost wall of the reactor. Most of the neutrons produced are absorbed in the blanket; it is here that tritium is produced and neutron energy is deposited.

Several classes of breeding materials have been considered:

- pure lithium metal with isotopic enrichments
- lithium alloys, including intermetallic compounds ( $\text{LiAl}$ ,  $\text{Li}_2\text{Sn}$ , and  $\text{Li}_7\text{Pb}_2$ )
- molten salts ( $\text{Li}_2\text{BeF}_4$  and  $\text{Li}_2\text{O}_3/\text{Li}_2\text{O}$  eutectic)
- solid ceramics ( $\text{LiAlO}_2$ ,  $\text{Li}_2\text{O}$ ,  $\text{Li}_2\text{C}_2$ ,  $\text{Li}_3\text{N}$ , and  $\text{LiD}$ ).

These materials may be used singly or in combination, with or without neutron multipliers and reflectors; their effectiveness depends on the design of the reactor.

In general, as atoms other than neutron multipliers are added to lithium to form compounds or alloys, the breeding ability of the combination decreases. Thus,  $\text{LiAl}$  is a poorer breeder than pure lithium, which in turn is poorer than  $\text{Li}_7\text{Pb}_2$ , where lead is a neutron multiplier.

## RECOVERING TRITIUM

The recovery rate of tritium from the breeding material usually depends on the concentration of tritium in the blanket. The concentration must climb to its steady-state value before recovery can proceed at a steady-state rate. The various breeding materials show vastly different behaviors with respect to the release of tritium.

The physical form of breeding materials is expected to be either powder or liquid so that processing kinetics will be fast.

There are three major recovery processes for tritium:

- permeation,
- trapping by adsorption, and
- trapping by absorption.

Molten salt extraction from liquid lithium followed by gas sparging may also be feasible and is being investigated.

In the permeation recovery process, tritium, in liquid lithium or high-pressure helium coolants, contacts one side of a selectively permeable membrane, such as niobium. On the other side of the membrane, the tritium partial pressure is lower so the tritium permeates the membrane. The rate of this continuous process is usually  $R_1 = K_1 (P_1^{1/2} - P_2^{1/2})$ , where  $K_1$  is a constant and  $P_1$  and  $P_2$  are the tritium partial pressures on the two sides of the membrane. Available evidence shows that when the permeation rate is surface limited it is better described by  $R_1 = V_2(P_1 - P_2)$ . In either case, the recovery rate increases as  $P_1$  (or the blanket tritium inventory) increases and is zero at startup.

The trapping processes generally operate by batch and require regeneration of the trap absorbents and adsorbents. Tritium can be absorbed from a vacuum or flowing helium stream by contact with a metal to form a stable hydride (typically cerium, scandium, or titanium).

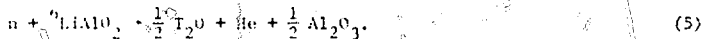
Tritium or tritiated vapors (water and hydrocarbons) can be removed from flowing gases by selective adsorbent filters, such as molecular sieves, silica gel, or activated charcoal.

In both trapping processes, first-order kinetics require the recovery rate  $R$  to be proportional to  $(P_1 - P_e)$ , where  $P_1$  is the partial pressure of the tritium-bearing species in the gas and  $P_e$  is the equilibrium partial pressure of the species being trapped at that particular temperature. Again, the recovery rate depends strongly on  $P_1$ , which is the principal factor in determining the blanket's tritium inventory.

Recovery of tritium from liquid lithium is expected to be difficult when the lithium concentration is kept below 10 ppm by weight. For this concentration, the blanket inventory would be about 5 kg (\$56,000,000) but the tritium partial pressure available to drive a separation process would be only  $10^{-10}$  torr at 1000 K, so large processing equipment might be required.

Adding aluminum to the lithium substantially raises the partial pressure<sup>6</sup>; this is the alloy in which tritium has been produced for defense purposes for many years.

An encouraging recent development is the renewed activity with  $\text{LiAlO}_2$  binders.<sup>7</sup> The tritium breeding reaction is



Tritium and tritiated water show a low solubility in the solid powder. Particles given the equivalent of a 1-h exposure to the neutron flux of a fusion reactor have released tritium in a mean time of 4.3 h at 600°C.<sup>6</sup> At high temperatures, it appears that tritium blanket inventories could be kept to a few grams in a large power plant. There are a few problems and unanswered

questions about oxide breeding materials, however. The high temperatures required to release the tritiated water may make necessary the use of refractory metals or carbides in structural members, but these materials are chemically attacked by water vapor at high temperatures. Prolonged radiation may induce swelling and internal void formation that could act as permanent sinks for a much larger amount of tritium than presently expected. These problems are common to all fusion reactor concepts and are independent of the plasma confinement concept.

#### CONTAINMENT OF TRITIUM

Environmental control of tritium in fusion reactors also presents a challenging set of problems. Tritium behaves just like hydrogen: it permeates metals and plastics, readily becomes airborne, and once mixed with normal water, it cannot be conveniently separated. The high tritium concentrations or partial pressures that provide high recovery rates from the breeding blanket may also cause high rates of permeation loss to the environment. For example, the reference design of the theta-pinch reactor calls for a liquid, lithium metal breeding blanket. To keep permeation losses to less than 30 Ci/d, the partial pressure of tritium has to be no more than  $10^{-10}$  Torr above the lithium.<sup>8</sup>

An obvious solution to some permeation losses is to use hydrogen-impermeable heat exchangers. This can be done by dropping the coolant operating temperature (and efficiency) or by adding new materials to the heat exchanger. Some of these new materials are barrier metals (copper, tungsten),<sup>8</sup> barrier oxides (oxidation films, BeO), and metal getters (titanium, cerium) and all might be placed into laminated (triplex) tubing to reduce or eliminate permeation losses.

High-level tritium in waste water is usually immobilized by reacting it to form concrete, which can then be encapsulated and buried. In the future, it may be possible to selectively remove the HTO from waste water by laser isotope separation techniques and greatly reduce the amount of solid waste to be buried.

Tritium in gaseous wastes can be reduced to low levels with the trapping processes used for recovering tritium from the breeding blanket. Reduction to the mpc has not yet been done economically. Tritiated hydrocarbons may be adsorbed directly or oxidized to trap the tritium as tritiated water on molecular sieves. This oxidation would normally be done at a few hundred degrees Celsius in an oxygen-rich stream flowing over precious metal catalysts. The gaseous species HT, DT, or T<sub>2</sub> can be trapped directly by adsorption or absorption. Usually, however, they are catalytically converted to water, or reacted with certain transition metal oxides (CuO, MnO<sub>2</sub>) to form water. Tritiated water is then trapped on molecular sieves. The removed fraction of tritiated water can be increased by deliberately swamping with water vapor after the first trapping, but this adds greatly to the liquid waste disposal problem. Clear trade-offs can and will be made strictly on an economic basis. Again, these problems are common to all fusion reactor concepts.

#### FUEL RECYCLING

Recycling the fuel in a fusion reactor is necessary because most of the fuel does not react before it is exhausted from the plasma region. Recycling problems are more specific to the reactor concept than those discussed so far. Mirror reactors are steady state in operation and have continuous neutral beam injection and exhaust. Injection requires separate deuterium and tritium streams. Tokamaks operate in a pulsed mode (about a 1-min cycle) with injection and between-cycle exhaust. The theta pinch is a short-pulsed reactor



(of a few seconds per pulse) and mixed isotopes in the fuel are desirable. Finally, laser-driven microexplosions are very short pulses (milliseconds). Fueling for these microexplosions is not even conceptualized: all that can be said is that the laser targets might be multiple, concentric spherical shells of frozen DT.

The recycle loop is fairly simple. Fuel may be introduced as gas at subatmospheric pressure or as energetic neutral particles and is exhausted through vacuum pumps. In a mirror reactor, the fuel exhausts directly to an electrical direct converter that recovers most of the kinetic energy of the escaping particles.

Several compounds must be removed from the fuel before it can be reused:

- helium ash,
- hydrogen from the  $D(T,n)He$  reaction (which occurs to a smaller extent than the normal  $D(T,n)He$  reaction), and
- methane.

Neutral beam injection allows preheated (high-energy) particles to be fed into the plasma across magnetic field lines. Some charge exchange reactions that must occur are velocity- rather than energy-dependent, so isotopic purity increases the efficiency of neutral beam injection. The separation of deuterium and tritium (along with the removal of hydrogen) will undoubtedly be accomplished with cryogenic distillation, an existing technology. Purely physical operations such as distillation must be supplemented with chemical methods to break down the DT, HD, and HT molecules. This is routinely done, but efficiency could be improved.

The innermost reactor wall may be lined on the plasma side with a graphite curtain to reduce neutron radiation damage. In mirror reactors, the direct-converter collection electrodes may also be graphite because of its high temperature properties. Energetic particles escaping from the plasma will be

implanted in the curtain materials. We suggest that a certain amount of the implanted tritium will be lost from the fuel stream because of chemical interaction with the curtain materials. This poses another unexplored inventory problem.

Cryogenic vacuum pumping seems to be straightforward if self-radiolysis and helium production from radioactive decay are not problems. Recent calculations suggest that self-heating is unimportant. Other pumping modes that will probably be used at various parts of the power plant include cryosorption, diffusion, and mechanical pumping. In these latter two pumping modes it is desirable to avoid hydrocarbon pump oils, but beyond that, there seems to be relatively little developmental work to be done.

In low-pressure plumbing systems, tritium reacts with hydrocarbons and other carbon compounds to form methane by an unknown, radiolytic mechanism. The presence of methane in the fuel supply is undesirable because it can plug the cryogenic still needed for hydrogen isotope separation, and can introduce impurities into the plasma. Whether additional methane will be formed by energetic implanted particles reacting with graphite is unknown. The cleanliness of vacuum system surfaces necessary to keep the plasma clean is a serious problem, particularly in Tokamaks, because impurities continuously accumulate during the relatively long cycle. Soviet workers recently estimated that to provide the low outgassing rates of the niobium first wall in their ETR Tokamak, the reactor should be outgassed at 2000 K for 4 days before operation.<sup>9</sup>

#### CONCLUSIONS

Fuel cycle problems in fusion reactors include the breeding, recovery, containment, and recycling of tritium. Technically, these problems are solvable; economically, their feasibility is not yet known.

REFERENCES

1. A. A. Moghissi and M. W. Carter, Eds., Tritium, (Messenger Graphics Phoenix, 1971).
2. R. L. Dobson, Low-Level Chronic Exposure to Tritium: An Improved Basis for Hazard Evaluation, Lawrence Livermore Laboratory Rept. UCRL-77372 (1975).
3. E. A. Evans, Tritium and Its Compounds, (John Wiley & Sons, New York, 1974).
4. D. Kramer, K. R. Carr, A. C. Pard, and C. G. Rhodes, A Survey of Helium Embrittlement of Various Alloy Types, Atomic International, Rept. AI-NEC-13047 (1972).
5. V. A. Maroni, et al., Liquid Metals Chemistry and Tritium Control Technology Annual Report, July, 1974-June, 1975, Argonne National Laboratory, Rept. ANL-75-50 (1975).
6. J. R. Powell, R. H. Wiswall, and E. Wirsing, Tritium Recovery from Fusion Blankets Using Solid Lithium Compounds, Brookhaven National Laboratory, Rept. BNL-23563 (1975).
7. A. A. Kishbaugh, Extraction of Tritium from Lithium Aluminate Targets, Savannah River Laboratory, Rept. DP-1058 (1966).
8. J. E. Draley, et al., An Engineering Design Study of a Reference Theta-Pinch Reactor (RTPR): Environmental Impact Study, Los Alamos Scientific Laboratory and Argonne National Laboratory, Repts. LA-5336, Vol. II, and ANL-8019 (1975).
9. Y. P. Vakhruzhin, et al., Planning Parameters for the ETRT Vacuum System: Preliminary Considerations, W. J. Grimes and Co., translators, Washington, D.C. (1974), ERDA-TR-17, Part 6.

MEMORANDUM FOR THE LOCAL REPRESENTATIVES  
AT THE 1976 ANNUAL MEETING,  
FEBRUARY 22 THROUGH 25, 1976, LAS VEGAS, NEVADA

☐ **FUEL CYCLE PROBLEMS IN FUSION REACTORS  
EVOLVE AROUND TRITIUM AND ITS**

- **B reeding, and recovery,**
- **Containment, and**
- **R ecycling.**

Fig. 1

☐ **TRITIUM:**

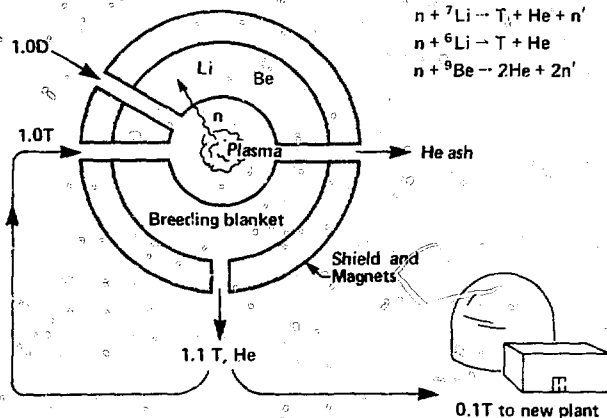
- **Is a required fuel for fusion reactors**
- **Easily undergoes fusion;  $T + D \rightarrow {}^4\text{He} + n + 17.6 \text{ MeV}$**
- **Decays spontaneously;  $T \rightarrow {}^3\text{He} + e + 5.6 \text{ keV}$**
- **Has a half-life of 12.3 years**
- **Occurs naturally in limited amounts and must be bred for fusion purposes**

☐ TRITIUM IS A HAZARD TO MAN AND HIS ENVIRONMENT:

- Like hydrogen, tritium may permeate, burn, and explode.
- It has a maximum permissible concentration of 1 ppt in air.
- One small drop of  $T_2O$  can be lethal.
- $T_2O$  is easily absorbed through the skin or lungs.
- It can cause hydrogen embrittlement.
- It can cause helium embrittlement by radioactive decay.

Fig. 3

☐ TRITIUM IS USUALLY BRED IN NUCLEAR REACTORS



Hickman - Fig. 4

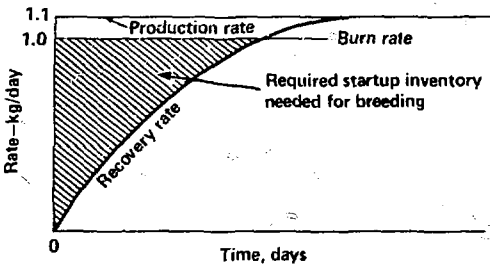
SEVERAL CLASSES OF BREEDING MATERIALS HAVE BEEN CONSIDERED:

- Pure lithium metal with isotopic enrichments;
- Lithium alloys, including intermetallic compounds, such as LiAl, Li<sub>2</sub>Sn, and Li<sub>2</sub>Pb<sub>2</sub>;
- Molten salts, such as Li<sub>2</sub>BeF<sub>4</sub> and LiNO<sub>3</sub>/LiNO<sub>2</sub> eutectic; and
- Solid ceramics, such as LiAlO<sub>2</sub>, Li<sub>2</sub>O, Li<sub>2</sub>C<sub>2</sub>, Li<sub>3</sub>N, and LiD.

These materials may be used singly or in combination, with or without neutron multipliers and reflectors; effectiveness depends on reactor design.

Fig. 5

BEFORE  $T_{out}/T_{in} = 1.1$ , THE TRITIUM CONTENT MUST CLIMB TO AN OPERATING STEADY-STATE VALUE BECAUSE ITS RECOVERY RATE DEPENDS ON CONCENTRATION



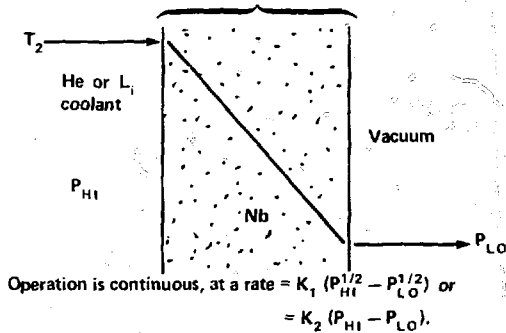
Bickman - Fig. 6

**RECOVERY PROCESSES**

- Permeation
- Adsorption trapping
- Absorption trapping
- Molten salt extraction

Fig. 7

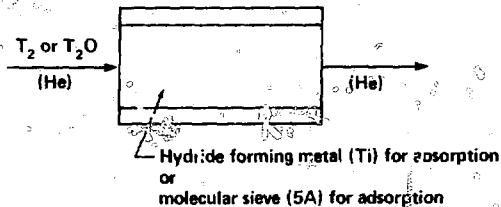
**PERMEATION RECOVERY PROCESS**  
Selectively permeable membrane



Hickman - Fig. 8



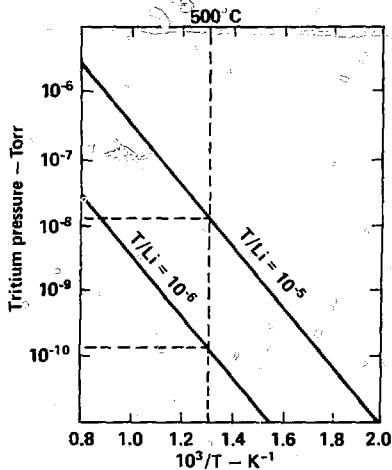
**ADSORPTION AND ABSORPTION-TRAPPING RECOVERY PROCESSES**



Operation is by batch, at a rate =  $K_3 (P_{H_1} - P_{E_0})$  or ?

Fig. 9

**THE CHEMICAL INTERACTION BETWEEN LI AND T MAKES RECOVERY FROM PURE LITHIUM DIFFICULT**



With  $10^6$  kg Li, the equilibrium inventory of tritium at 500°C is  
0.5 kg < inventory < 5 kg if  
 $10^{-10} < P < 10^{-8}$  Torr is maintained

Hickman - Fig. 10

**ALLOYING LITHIUM WITH ALUMINUM GREATLY RAISES THE TRITIUM PARTIAL PRESSURE OVER LITHIUM BUT REDUCES THE BREEDING RATIO AND PROBABLY AGGRAVATES CORROSION**

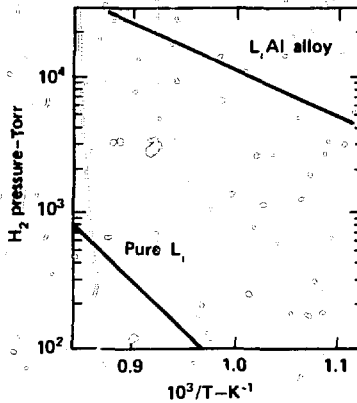
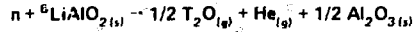


Fig. 11

**${}^6\text{LiAlO}_2$  WITH A NEUTRON MULTIPLIER LOOKS ATTRACTIVE AS A BREEDING MATERIAL BECAUSE IT PROMISES A VERY LOW BLANKET TRITIUM INVENTORY**



Inventory for 1000 Mw<sub>e</sub> = 1 g T<sub>2</sub>!

Concentration > 5 ppb

**${}^6\text{LiAlO}_2$  POWDER COULD BE USED IN THE BREEDING REGION**

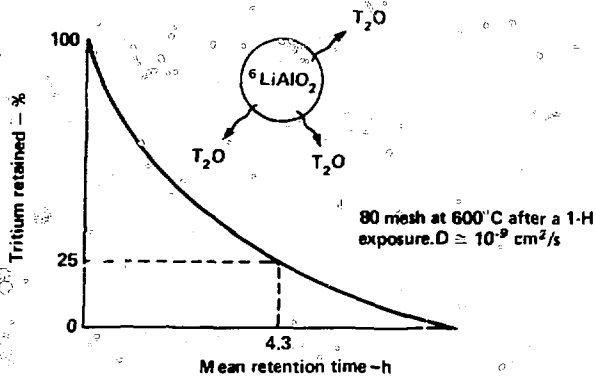


Fig. 13

**POTENTIAL PROBLEMS OF  $\text{LiAlO}_2$**

- $\text{T}_2\text{O}$  release requires high-temperature operation
  - High temperature structure materials are niobium, vanadium, molybdenum, and carbon
  - $\text{T}_2\text{O}$  corrodes these materials
- Radiation swelling and internal bubble formation may trap tritium in the ceramic

**CONTAINMENT (ENVIRONMENTAL CONTROL)**

Difficult because  $T_2$  behaves like normal hydrogen, readily converting to water and exchanging with hydrogen in hydrocarbons

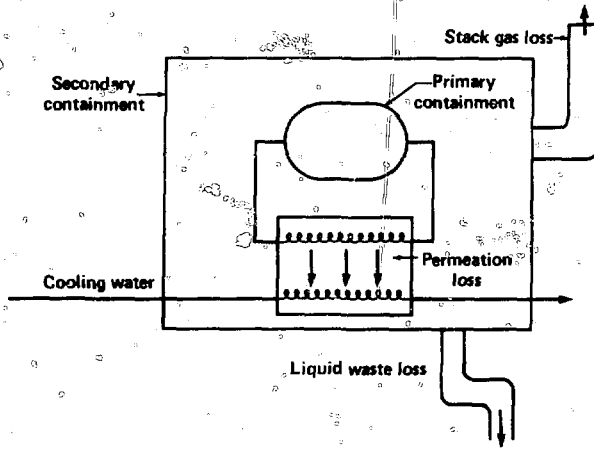


Fig. 15

**HIGH CONCENTRATIONS (PARTIAL PRESSURES) THAT PROVIDE HIGH RECOVERY RATES MAY CAUSE HIGH RATES OF PERMEATION LOSS TO THE ENVIRONMENT**

**Example:**

The theta pinch reference design calls for a Li metal breeder and a  $T_2$  partial pressure of  $10^{-10}$  torr to keep permeation losses below 30 Ci/d.

**AN OBVIOUS SOLUTION TO SOME PERMEATION LOSSES IS TO USE HYDROGEN-IMPERMEABLE HEAT EXCHANGERS**

- Drop temperature (and efficiency)
- Add new materials
  - Barrier metals (Cu, W)
  - Barrier oxides ( $\text{Cr}_2\text{O}_3$ , BeO)
  - Getter metals (Ti, Ce) at low temperature only

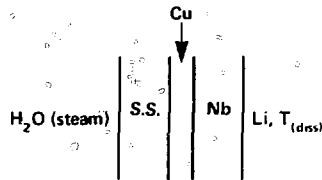
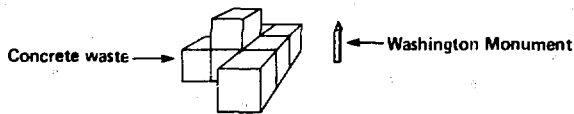
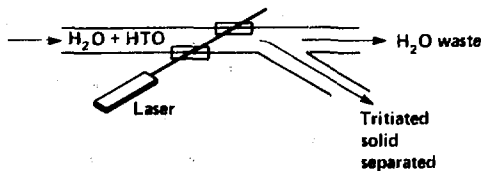


Fig. 17

**HTO LIQUID WASTE IS CONVERTED TO CONCRETE, ENCAPSULATED, AND BURIED**



Eventually, it may be possible to selectively remove the HTO from waste water by laser isotope separation.



Hickman - Fig. 18

## **RECYCLING**

**RECYCLING IS NECESSARY BECAUSE ALL FUEL IS NOT REACTED BEFORE BEING EXHAUSTED FROM THE PLASMA REGION. RECYCLING PROBLEMS ARE SPECIFIC TO EACH REACTOR DESIGN:**

- Mirror reactors have continuous injection of separate D and T streams and continuous exhaust
- Tokamaks have injection (pellets, beams), operate in a pulsed mode (of about on 1-minute cycles), and exhaust between cycles
- Theta pinch has a cyclic (several seconds) fuel injection and exhaust with mixed isotopes
- Laser-driven microexplosion is very short-pulse (milli-seconds) but is a separate technology.

Fig. 19

## **TRITIUM IN GASEOUS WASTES CAN BE REDUCED TO VERY LOW LEVELS, BUT NOT, ECONOMICALLY, TO THE MPC LEVEL**

- Tritiated hydrocarbons: adsorb directly or oxidize and trap HTO
- HT: adsorb or absorb directly on hydride-forming metals or oxidize and trap HTO. Oxidation:  $O_2$  on a Pt/Pd catalyst or react with  $CuO$ ,  $MnO_2$
- HTO: adsorb directly. Water aids adsorption but increases the quantity of liquid waste.

Current technological limit is about 1 ppb.

Hickman - Fig. 20

THE RECYCLE LOOP IS FAIRLY SIMPLE

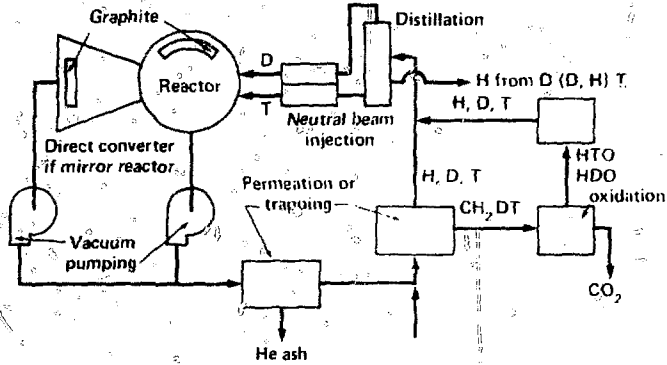


FIG. 21

NEUTRAL BEAM INJECTION ALLOWS INTRODUCTION TO THE REACTOR OF PREHEATED (HIGH ENERGY) PARTICLES ACROSS MAGNETIC FIELD LINES

But

Requires large-scale application of cryogenic distillation at 20K, (an existing process) and a high tritium inventory.

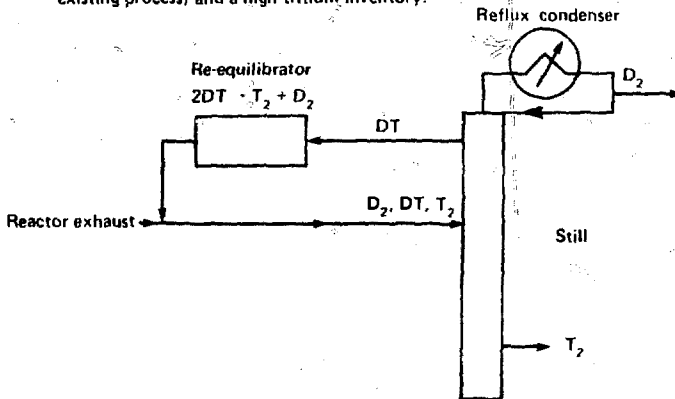


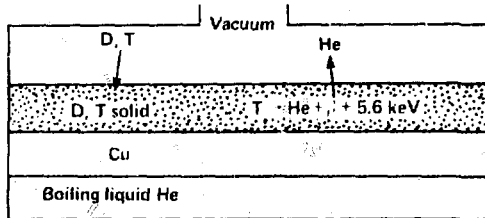
FIG. 22 - FIG. 23

A graphite lining in the plasma chamber will reduce neutron radiation damage.

Tritium will be implanted in the graphite and may be permanently trapped by a chemical reaction, which presents an unexplored inventory problem.

Fig. 23

CRYOGENIC VACUUM PUMPING SEEMS TO BE STRAIGHTFORWARD IF THE SELF-RADIOLYSIS AND HELIUM PRODUCTION DO NOT HAVE LARGE EFFECTS ON THE VACUUM. SELF-HEATING SEEMS NEGLIGIBLE



Other pumping options:  
Diffusion, cryosorption, turbomolecular pumps.

Rickman - Fig. 24



**IN LOW-PRESSURE PLUMBING SYSTEMS, TRITIUM REACTS WITH HYDROCARBONS AND OTHER CARBON COMPOUNDS TO FORM METHANE BY AN UNKNOWN RADIOLYTIC MECHANISM.**

- 1) Stili plugging
- 2) Plasma quenching

**This is more important for Tokamaks because impurities accumulate for the duration of the burn. Soviet workers envision an initial, four-day bake-out at 2000K to clean the niobium first wall!**

Fig. 25

## **CONCLUSIONS**

**Fuel cycle problems in fusion reactors include the breeding, recovery, containment, and recycling of tritium. Technically, these problems are solvable; economically, their feasibility is not yet known.**

Hickman - Fig. 26