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TRITIUM HANDLING IN VACUUM SYSTEMS

A Short Course Offered by
The American Vacuum Society
Baltimore, Maryland

16 November 1982

UPDATED OCTOBER 1986

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August 1982
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I. PROPERTIES OF TRITIUM

J. T. GILL

I.A. CHEMICAL PROPERTIES OF TRITIUM

I.A.1 HYDROGEN ISOTOPES

a. There are three well-known isotopes of hydrogen:

Symbol	Name	Abundance	Mass
${}_1\text{H}^1(\text{H})$	Protium	99.985	1.008142
${}_1\text{H}^2(\text{D})$	Deuterium	0.015	2.014735
${}_1\text{H}^3(\text{T})$	Tritium	β -Emitter 12.32 yr half-life	3.016997

${}_1\text{H}^4$ and ${}_1\text{H}^5$ are also known, but decay in fractions of 1 sec, rendering them relatively useless.

b. Several isotopic combinations of simple molecules can be formed:

- Hydrogen molecules (6 combinations):

- Water (6 combinations):

- Methane (12 combinations):

- Hydrogen Fluoride (3 combinations):

c. Tritium is most often handled, in vacuum systems at room temperature, as the molecular species T_2 .

I.A.2 AT LOW CONCENTRATION, TRITIUM REACTS WITH COMPOUNDS AS
WOULD H OR D:

SUBSTITUTION OR EXCHANGE REACTIONS, WHICH ARE RAPID FOR
H OR D, ARE ALSO RAPID FOR TRITIUM:

I.A.3 THE RATES/EQUILIBRIUM DISTRIBUTIONS OF REACTIONS INVOLVING HYDROGEN ARE GENERALLY ISOTOPE DEPENDENT.

- a. This is particularly so if the bond involving the hydrogen atom in question is broken during the reaction (or involved in transition state):

$$(\text{C}_6\text{H}_6/\text{C}_6\text{H}_5\text{T}) \sim 2.5\% \text{ different}$$

T to P

By contrast:

- b. Another example:

$$(k_H/k_D)_{25^\circ\text{C}} = 6$$

c. Self-ionization of water (acidities):

d. Presence or absence of isotope effects are often used by chemists to support or refute proposed reaction mechanisms.

e. Most hydrogen isotope effects are within factors of 10-20.

f. Effects caused by differences in zero-point energies (hence bond dissociation energies required) in compounds:

The less energy required to dissociate a bond (or drive that bond to a length required in a transition state to a new compound),

the faster the reaction.

I.A.4 THERE IS A DIFFERENT, ADDITIONAL ISOTOPE EFFECT TO BE CONSIDERED IN THE CASE OF TRITIUM REACTIONS.

- a. Many reactions are thermodynamically favored but kinetically slow for H or D species:

heat of reaction = -16 kcal/mole (CH_4 favored)

but half-life for reaction at 25°C ~ years.

- Activation energy necessary to drive reaction is not present at room temperature.

- b. With tritium, however, activation energy is present as β -rays (high energy electrons). Upon collision with molecule, β will lose several eV (1 eV = 23 kcal/mole) and produce dissociated molecular fragments, ions, radicals.

Half-life for reaction (τ) at 25°C ~ hours.

T_2 is a "self-catalyzing reducer."

c. Other reactions proceed concurrently with

depending upon relative proportions of reactants.

- If $[\text{C}_2\text{H}_6] \gg [\text{T}_2]$:

Most β energy is deposited in ethane, forming organic radicals and ions which, in turn, react primarily with ethane. Higher order hydrocarbons and polymers, containing some T, are formed.

- If $[\text{C}_2\text{H}_6] \ll [\text{T}_2]$:

Most β energy is deposited in T_2 , forming T radicals and ions which will reduce C_2H_6 to CT_4 , protium being redistributed between CHT_3 and HT.

- Between these extremes, above reactions will all occur in varying proportions.

d. Another example:

$\frac{d[DT]}{dt} \propto [T_2]/([D_2] + [T_2])$. Probably the creation of reactive species (atoms, ions, electrons) is the rate determining step. Those species are formed in both D_2 and T_2 , but are generated from T_2 β -radiation.

I.A.5 BECAUSE OF ITS RADIOACTIVE, CHEMICALLY REDUCING, AND DIFFUSIVE PROPERTIES (SEE BELOW) TRITIUM GAS (1 ATM.) INTERACTS IN SOME WAY WITH NEARLY ALL MATERIALS.

- a. Organics, e.g., pump oils, plastics, o-rings: T_2 will perfuse and degrade many useful polymeric materials, causing loss of mechanical properties within days-months.
- b. Glasses: T_2 will diffuse through glasses; β will activate the reduction of Si-O-Si bonds to Si-OT and Si-T bonds; mechanical properties will be lost, but only after years.
- c. Ceramics: Situation similar to glasses, but slower diffusion; reduction of network bonds is again likely; mechanical properties are lost only after many years.
- d. Metals: Some metals, e.g., uranium, are directly "hydrided," forming a chemical compound and changing mechanical properties grossly in minutes-hours. Other metals, e.g., stainless steels, are permeated by T_2 (a problem in itself) but do not lose mechanical properties unless T_2 pressure is hundreds of atmospheres (strength lost in ~5 years).

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I.B. NUCLEAR PROPERTIES OF TRITIUM

I.B.1 FOR THE DISINTEGRATION OF TRITIUM:

HALF-LIFE IS 12.32 YEARS; DISINTEGRATION PROBABILITY

IS $\frac{1.78234}{1.783} \times 10^{-9} \text{ sec}^{-1}$.

a. $({}_2\text{He}^3)^+$ formed in tritium decay is rapidly neutralized by thermal electrons; ${}_2\text{He}^3$ is a stable isotope of helium.

b. Since 1 Curie = 3.700×10^{10} disintegrations/sec,
1 cm³ of T₂ gas at 1 atm (0°C) = 2.59 Ci, (25°C) = 2.37 Ci;
1 g = ~~9619~~ Ci.

9616.9 C.

I.B.2 THE β -SPECTRUM OF TRITIUM DECAY, NUMBER DENSITY VERSUS ENERGY OF β PARTICLE, IS SHOWN ON THE NEXT PAGE.

FROM THIS AND OTHER DETERMINATIONS, THE FOLLOWING β ENERGY PARAMETERS ARE ACCEPTED:

$$E_{\max}(\beta) = 18.5 \text{ keV}$$

$$\bar{E}(\beta) = 5.69 \text{ keV}$$

III. Investigation of Soft Radiations—II. The Beta Spectrum of Tritium*

Phil. Mag. 40, 53 (1949)

By S. C. CURRAN, J. ANGUS, and A. L. COCKROFT,

Detail of spectrum in the low energy region.

I.B.3 ENERGIES OF RECOIL FRAGMENTS

- a. Upon beta-emission the remaining (He^3 -containing) molecular fragment will recoil with a kinetic energy of E_R , where:

$$E_R = E_\beta (M_\beta + M_0) / 2 M_R$$

E_β is the β particle energy

M_β is the relativistic mass of the β particle

M_0 is the rest mass of the β particle

M_R is the mass of the recoil fragment.

- b. Since $M_\beta / M_0 = 1.04$ for the highest energy tritium particle the above simplifies to:

$$E_R \cong E_\beta M_0 / M_R = 5.5 \times 10^{-4} E_\beta / M_R',$$

M_R' is expressed in atomic weight units.

- c. For the reaction

If $E_\beta = 18.5$ keV, then $E_R(\text{He}^3)^+ = 3.4$ eV.

d. For $\text{HT} \rightarrow (\text{HHe}^3)^+ + \beta^-$:

If $E_\beta = 5.7 \text{ keV}$, then $E_R(\text{HHe}^3)^+ = 0.78 \text{ eV}$.

Since bond energy of $(\text{HHe}^3)^+$ is $\sim 2 \text{ eV}$, the recoil kinetic energy is not enough to cause dissociation of the molecule. $(\text{HHe}^3)^+$ remains bound until it combines with an electron or undergoes collision:

I.B.4 THE LOSS OF ENERGY OF A β -PARTICLE BY COLLISIONS IN A PARTICULAR MEDIUM IS A FUNCTION OF ENERGY:

β Energy	$-\frac{dE}{dx}$ (Linear Energy Transfer) in H_2O
0.01 MeV	0.232 eV/Å
0.1	0.042
1.	0.018
10.	0.020

a. For non-relativistic electrons

$$LET = -\frac{dE}{dx} = \frac{C\rho Z}{N} \cdot \frac{\ln(f(z) \cdot E)}{E}$$

C = constant

ρ = density of medium

Z = atomic number (e^- /atom) of medium

N = atomic weight of medium.

b. Because of this known rate of energy loss, the range of a β should also be known. Empirically, for $E(\beta) < 2.5$ MeV, range R_{Al} in aluminum:

$$R_{Al} = 412E^x, \quad x = 1.265 - 0.094 \ln E.$$

A fit of data (from different β -emitters) to this equation is shown on the next page.

c. Range-energy curve for β -particles in aluminum

d. For a 5.7 keV β (average tritium β energy),

$$R_{Al} = 0.049 \text{ mg/cm}^2.$$

$$\text{Range (cm) in aluminum} = R' = \frac{R_{Al}}{\rho} = \frac{0.049 \text{ mg/cm}^2}{2.7 \times 10^3 \text{ mg/cm}^3} = 1.8 \times 10^{-5} \text{ cm}$$

- e. To get range in another material, one must correct for relative scattering power per unit weight. Since scattering is primarily from electrons in material:

$$\text{Range} = R' = (R_{Al}/\rho) \frac{N \cdot 13}{Z \cdot 27} \quad \begin{array}{l} N = \text{atomic weight} \\ Z = \text{atomic number} \end{array}$$

- f. So range of 5.7 keV β in T_2 gas at 1 atm:

$$R' = \frac{0.049}{6/22.4} \cdot \frac{6 \cdot 13}{2 \cdot 27} = 0.26 \text{ cm.}$$

- g. Maximum range of β in T_2 gas at 1 atm:

$$R' = 3.2 \text{ cm.}$$

- h. Absorbance of β energy with distance follows closely the equation:

Fraction absorbed = $1 - \exp(-\mu x)$, where

x = distance

μ = absorption coefficient

= 1.81 cm^{-1} for H_2 at 1 atm, 25°C

11.0 N_2

12.9 Ar

$\mu \propto$ # electrons/cc for a material

Example: At 3.2 cm of H_2 gas, 1 atm, 25°C :

absorbed energy = 99.7%.

I.B.5 WHEN A β PARTICLE ENTERS A MATERIAL, SEVERAL PROCESSES OCCUR UPON COLLISION OF THE "HOT" ELECTRON WITH THE ATOMS/MOLECULES.

a. Free radicals are generated upon bond scission:

These radicals will recombine or transmute, generating original or altered species.

b. Ions and ion-radicals are generated upon promotion of electrons above bound states:

These species will also further react, often forming altered bonding arrangements.

c. In the reaction mentioned earlier,

All of the following processes can occur:

plus all $(D_m T_n)^+ + e^- \rightarrow D_m T_n$ recombinations.

This is a relatively simple overall process, but even here the variety of initiating and subsequent reactions is large.

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I.C. INTRODUCTION TO BIOLOGICAL PROPERTIES OF TRITIUM

I.C.1 TRITIUM IS BIOLOGICALLY DAMAGING AS WELL, THOUGH NOT SO BAD AS HIGHER ENERGY RADIONUCLIDES.

- a. No specific organs of concentration in the body: distribution in body water. Dose is ~homogeneous throughout body.
- b. Uptake generally through lungs or hands; homogeneous distribution in body after ~90 minutes.
- c. After large uptake, only 1-2% of T is organically bound. During chronic exposure 25-40% becomes organic, due to enzymic reactions within body.
- d. No deaths or severe illnesses from acute exposure have occurred.
- e. Disruption of cellular DNA from β -particle induced recoil, radical, and ion reactions is the long-term worry from chronic exposure.

I.C.2 BECAUSE BODY WATER IS CONTINUALLY BEING RECYCLED, DOSE FROM A SINGLE ACUTE EXPOSURE IS LIMITED.

a. Body half-life of water supply (and hence most of tritium) is 8-12 days, depending upon fluid intake.

b. When exposed, drink!!

c. Tritium uptake monitored via routine analysis of urine samples.

d. Dose calculated from urine count data.

e. Actual example:

<u>Date</u>	<u>Count(μCi/l)</u>
1/29	0
2/8	Exposure! count unknown
2/13	4
2/19	1
2/20	1
2/25	0

count
($\mu\text{Ci}/\ell$)

- f. Total exposure calculated from:
Initial count (extrapolated, if necessary)
 $\times$
 $4 \text{ mrem} \cdot \ell / \mu\text{Ci}$ (population average proportionality constant)
- g. So the person received:
 $\sim 8 \times 4 = 32 \text{ mrem total dose.}$

I.C.3 EXPOSURE FROM TRITIATED WATER (HTO OR T_2O) IS $\sim 10^4$ MORE DANGEROUS THAN FROM TRITIATED HYDROGEN (HT OR T_2).

a. Kinetics of exchange:

much faster than:

b. In addition, the progress of the reaction

is slow at low T_2 concentration in air without catalyst (half-life $\sim$ months).

I.C.3.b Rate constant for $T_2 + O_2 \xrightarrow{k} T_2O + O$

$$\frac{[T_2O]_t}{[T_2]_0} = [1 - \exp(-kt)]$$

c. So the release from the previous section, in the form of $\sim 10 \text{ Ci } T_2$ gas, resulted in a small dose to the operator. Had the release been HTO

I.C.4 EXPOSURE LIMITS TO TRITIUM ARE RECOMMENDED BY THE INTERNATIONAL COMMISSION OF RADIOLOGICAL PROTECTION (ICRP).

- a. Annual uptake limit = 5 rem $\sim$ 1200 μ Ci/l $\sim$ 81 mCi
Mound guideline = 0.75 rem
- b. Derived permissible air concentration
= 2×10^{-5} Ci/m³ (HTO)
= 0.5 Ci/m³ (HT).
- c. Urine counts $\leq$ 10 μ Ci/l will be tolerated. Above 50 μ Ci/l worker is removed from area until counts are down.

I.C.4.d From the code of U.S. Federal regulations:

RADIOACTIVE CONCENTRATION GUIDES
FOR TRITIUM

	<u>AIR</u>	<u>WATER</u>
Occupational for 40 HR Week *		
Gas	$2 \times 10^{-3} \mu\text{Ci/cc}$	
Oxide	$5 \times 10^{-6} \mu\text{Ci/cc}$	$1 \times 10^{-1} \mu\text{Ci/cc}$
Off-Site for Individuals		
Gas	$4 \times 10^{-5} \mu\text{Ci/cc}$	
Oxide	$2 \times 10^{-7} \mu\text{Ci/cc}$	$3 \times 10^{-3} \mu\text{Ci/cc}$
Off-Site for General Population		
Gas	$1.3 \times 10^{-5} \mu\text{Ci/cc}$	
Oxide	$7 \times 10^{-8} \mu\text{Ci/cc}$	$1 \times 10^{-3} \mu\text{Ci/cc}$

*Uses factor of 400 for comparative $\frac{\text{HTO}}{\text{HT}}$ hazard.
Other sources quote factor of ~25,000, allowing
very loose HT standard.

I.C.4.e Philosophy for setting exposure limits

In determining biological effects, it is useful to correlate the dose received with the response or damage produced. In plotting these two variables, a dose-response-curve can be drawn. An important question has been the shape of this curve. I will illustrate two possibilities.

Most effects fall into the category of threshold effects. That is, a certain minimum dose must be exceeded before an effect is observed. This is a typical threshold dose-response curve.

Non-threshold effects, as the name suggests, means that any radiation dose, no matter how small, involves some degree of biological effect. For non-threshold effects, a linear dose-response curve is obtained.

Genetic effects of radiation are considered to be a non-threshold phenomenon.

Radiation protection guides for large populations are established based on this concept.

The objectives of radiation protection are to prevent somatic effects by setting dose limits at sufficiently low values so that no threshold dose would be reached, and to limit the probability of genetic effects to levels deemed acceptable.

- I.C.5 HYPOTHETICALLY, ASSUME A TRITIUM GAS HANDLING SYSTEM CONTAINS T_2 GAS AT 1 ATM AND HAS DEVELOPED A LEAK WHICH ALLOWS T_2 ESCAPE OF 1×10^{-3} cc/sec.
- a. Assume the leak is near a hot surface, an active metal surface, or an electric motor.
 - b. Assume therefore that 10% of T_2 is converted to HTO .
 - c. Assume the operator is nearby for 4 hours and he/she respirates 10% of HTO .
 - d. Uptake will be:
 $1 \times 10^{-3} \times 2.6 \times 60 \times 60 \times 4 \times 0.1 \times 0.1 = 360 \text{ mCi},$
well in excess of annual limit.

I.C.6 IF, HOWEVER, A VENTILATION OR CONTAINMENT SYSTEM IS PRESENT WHICH SWEEPS ALL BUT 1/1000 OF THE LEAKING TRITIUM AWAY (TO STACK OR TO RECYCLE), UPTAKE WILL BE . 0.360 mCi ~ 6 μ Ci/l.

- a. A reasonably small exposure. Ventilation or containment of T_2 is important for tritium work.
- b. Fume hoods or, better yet, gloveboxes with service from a tritium cleanup system are standard for housing of major tritium systems. See next page.

I.C.7 Monitoring for Tritium

a. AIR (lab and glovebox): Ionization chambers

1. Sensitivity $\sim 1 \times 10^{-6} \text{ Ci/m}^3$
2. Portable and permanent units, γ compensation available:
 - M. Overhoff & Assoc., Cincinnati, OH
 - Wm. Johnson & Assoc., Montville, NJ
 - Ontario Hydro/Clarkson Site, Mississauga, ONT
3. Diagram of a typical (permanent) installation:

I.C.7.b SURFACES

1. Filter paper swipes
 - Rub over $\sim 200 \text{ cm}^2$ of surface
 - Place in proportional counter ($\sim 30\%$ efficiency)
2. Plastic scintillation device
 - For flat, opaque surfaces only
 - Hughes Whitlock Ltd. (via Overhoff)
3. Sensitivity of both ~ 10 disintegrations/min.
4. Qualitative methods only

c. PERSONNEL

1. Film badges not good for tritium
2. Filter paper swipes of skin during and after hot jobs
3. Urine count
 - 2-3 times per week if working actively in tritium operations
 - once/wk. if office work only
 - $\sim 20 \text{ ml}$ given
 - 1 ml sample mixed with 5 ml Atom Light scintillation cocktail, then counted for 1 min.; efficiency $\sim 30\%$
 - Sensitivity $\sim 0.02 \times 10^{-6} \text{ Ci/l}$

I.C.7.d ENVIRONMENTAL

1. On-site : stacks

- Ionization chambers
- Glycol bubblers (differentiating)
 - HTO in stack gas absorbed into ethylene glycol in first bubblers
 - HT in stack gas passes through first bubblers, is oxidized, then absorbed in second bubbler set
 - Glycol is periodically removed for measurement in scintillation counter
 - Sensitivity $\sim 1 \times 10^{-10} \text{ Ci/m}^3$
- Flow schematic of typical effluent monitoring system:

Flow schematic of Mound effluent monitoring systems.

- Flow schematic of differentiating glycol bubbler :

FIGURE 2 - Modified differentiating tritium sampler (1979).

2. Off-site

- Glycol bubblers (HTO only) situated in community up to ~50 Mi from site
- Water, soil, vegetation samples from environs
 - Scintillation counted
 - Several thousand per year

I.C.8 CONTAMINATION CONTROL : EQUIPMENT & METHODS

a. Operations

- Fumehoods, gloveboxes, pass chambers, bagging ports
- Daily surface swipes of operating areas
- Ion chamber monitoring of lab air (particularly around hoods & boxes), glovebox atmospheres

b. Maintenance

- Inside glovebox when possible
- Tents and airlocks otherwise
- Tenting flanges around glovebox windows

c. Emergency

1. Release of tritiated gas
 - Within glovebox : cleanup system
 - Into room: evacuation, then stacking or room cleanup system
2. Release of contaminated liquids or particulates: area isolation, decontamination

I.C.9 PROTECTIVE CLOTHING

a. Standard

1. Offices: smock (washable) over street clothes, plastic covers (disposable) over street shoes
2. Labs: "blues" (washable shirt and trousers), "hot" shoes (kept in radiation area), cotton and surgeon gloves (disposable) when handling contaminated material

b. Maintenance

1. Bubblesuits with shoe covers

**GLOVES OF VARIOUS PROPERTIES WERE EVALUATED
FOR USE IN BUBBLE SUIT JOBS**

<u>GLOVE MATERIAL</u>	<u>RELATIVE HTO PERMEATION RATE (FOR UNIT THICKNESS)</u>	<u>THICKNESS (MIL)</u>	<u>COLORING</u>	<u>TOUGHNESS</u>	<u>1973 COST PER PAIR</u>
NATURAL RUBBER	900	7 15	NATURAL ORANGE	LOW	\$0.21 0.32
NEOPRENE	300	15	BLACK	MEDIUM	1.03
BUTYL RUBBER	1	13	BLACK	HIGH (BUT AVOID ORGANICS)	2.25
BUNA-N	400	15	BLUE	VERY HIGH	0.90

**TWO CHOICES FOR GLOVE APPAREL WERE
CHOSEN FROM THE EVALUATIONS**

HTO JOBS:

COTTON, 7 MIL NATURAL, BUTYL,
BUNA-N, 15 MIL NATURAL
(SOME TACTILE SENSATION LOSS)

ORGANIC & OTHER JOBS:

COTTON, 7 MIL NATURAL (2X),
BUNA-N, 15 MIL NATURAL
(LITTLE TACTILE SENSATION LOSS)

I.C.10 MAINTENANCE JOBS

- a. Planning meeting (held weekly)
- b. Pre-job conference
- c. Work area isolation (tenting & bagging)
- d. Bubblesuit, health physics backup
- e. Dose record
- f. Decontamination

I.C.11 PACKAGING AND SHIPMENT OF TRITIUM

d. PACKAGING REQUIREMENTS FOR TRITIUM AS A GAS OR ABSORBED ON SOLID MATERIAL

CATEGORY	AMOUNT	CONTAINER	LABEL
LSA (waste category)	<0.3 mCi/g 136.08 mCi/lb 13,608 mCi/100 lb 0.13608 Ci/lb 13.608 Ci/100 lb 136,080 μ Ci/lb	Strong, leak-tight package • 17H or 17C metal drum with metal liner if out-gassing; lid sealed with RTV sealant • Standard wood box if no out-gassing • Welded steel box (type 7A) if out-gassing	LSA LABEL, RADIA- TION SYMBOL if sole-use vehicle if loaded by consignee if unloaded by consignor
SMALL QUANTITY	Up to 25 Ci/ pkg (<0.5 mrem/ hr at external surface of package)	Strong, leak-tight package	Outside of inner package labeled "RADIOACTIVE"
TYPE A	Up to 1000 Ci/ pkg	17H or 17C metal drum AL-M1, 6C drum Welded steel box (type 7A)	WHITE I LABEL (if not ex- ceeding 0.5 mrem/hr at ex- ternal surface)
TYPE B	1000 to 50,000 Ci/pkg	17H or 17C metal drum AL-M1, 6C drum Welded steel box (type 7A)	WHITE II LABEL (as above)
LARGE QUANTITY	>50,000 Ci/pkg	17H or 17C metal drum AL-M1, 6C drum Welded steel box (type 7A)	YELLOW III LABEL

1. Water from glovebox cleanup systems
 - 100-1000 Ci/l, depending on R.H.
 - Electrolyzed for recovery (shipped via AL-M1)
or sorbed onto concrete in drums (sealed & buried)
2. Oil from vacuum pumps, etc.
 - 20-40 Ci/l if aliphatic oil
 - Solidified onto inorganic sorbant in drums
(sealed & buried)
3. Contaminated hardware
 - > 50 c/m on surface swipes, Ci content only
roughly known
 - Breakdown to smallest size, place in drum or steel
box (sealed or welded, buried)

I.C.12 REFERENCES

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I.D. SOURCES OF TRITIUM

I.D.1 THERE ARE SEVERAL ENVIRONMENTAL SOURCES OF T.

a. Tritium occurs naturally in the environment.

1. Reactions of cosmic rays with upper atmosphere constituents produce tritium. Examples:

2. Tritium produced becomes incorporated in water and falls to earth as rain.

3. Estimated production of 4×10^6 Ci/year results in inventory of $\sim 70 \times 10^6$ Ci.

b. Nuclear test explosions from 1945 to 1975 contributed some additional 1900×10^6 Ci.

c. Commercial products contribute $\sim 1 \times 10^6$ Ci/year.

d. Nuclear Industry

1. Present releases $\sim 1 \times 10^6$ Ci/year.
2. Substantial pressures to decrease this value as low as practicable. Over the last ~ 12 years at Mound, airborne releases have been decreased by a factor of 100. See next page.
3. Depending on developments within the nuclear power industry, releases could reach 4×10^6 Ci/yr by 1990 and 50×10^6 Ci/yr by 2010.

TRITIUM RELEASED IN AIR

I.D.2 VARIOUS POTENTIAL AND ACTUAL SOURCES OF TRITIUM PRODUCTION
EXIST IN THE U. S. AND CANADA.

a. Light-water Reactors

1. ${}_0n^1$ capture in boron of primary coolant.
2. $\sim 500\text{--}1000$ Ci/yr per 1000 MW power.
3. May be 3×10^5 Ci/yr total by year 2000.
4. Low concentration - not economical to recover.

b. Heavy-water Reactors

1. ${}_0n^1$ activation of D_2O .
2. $\sim 2 \times 10^6$ Ci/yr per 1000 MW power.
3. May be 2×10^7 Ci/yr total by 1985,
 2×10^8 Ci/yr total by 2000.
4. Recovery at $\sim \$0.50/\text{Ci}$ envisioned.

c. Fuel Reprocessing Plants

1. Ternary fission product from fuel.
2. May reach 2×10^6 Ci/yr by 1985,
 1×10^7 Ci/yr by 2000, if reprocessing is allowed.
3. Recovery cost, a function of process, between
 $\$0.20 - 2.00/\text{Ci}$.

d. DOE Production Reactors

1. ${}_0n^1 + {}_3\text{Li}^6 \rightarrow {}_1\text{H}^3 + {}_2\text{He}^4$
2. Irradiation of Li/Al alloys or LiAlO_2 ceramic.
3. Hanford N reactor capable of 6×10^7 Ci/yr production via aluminates.
4. Other capabilities secret.
5. T_2 gas (>98%) available through Oak Ridge National Laboratory (commercial) or Savannah River Plant (DOE internal).
 - a) containers from 10 cm^3 to 50 liter,
 - b) \$2.00/Ci below 1000 Ci, and
 - c) \$0.75/Ci above 25,000 Ci.

I.D.3 THE ABOVE SOURCES WILL BE TAPPED FOR OPERATION OF FUTURE FUSION REACTORS AND EXPERIMENTAL SUBSYSTEMS.

- a. Tritium systems test assembly (1982):
 $\sim 2.5 \times 10^6$ Ci; experimental tritium recycle.
- b. Tokamak Fusion Test Reactor (~1985-9): $\sim 4 \times 10^5$ Ci/yr;
 non-recycle.
- c. Commercial (2010?): $2 \times 10^7 - 2 \times 10^8$ Ci/1000 Mw; for
 initial startup only; recycle and breeding.

I.D.4 REFERENCES

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I.E. PHYSICAL PROPERTIES OF TRITIUM

1. SOLUBILITY OF TRITIUM IN MATERIALS.

- a. H, D, and T dissolve to some extent in all materials.
 1. Isotope effects are small - see below.
 2. Great range in solubilities: solubility in elastomers $\sim 10^{13}$ greater than in tungsten at 1 atm, 300°K.
 3. Hydrogen atoms or molecules dissolve by diffusing into a structure and locating in sites between atoms or molecular frameworks - interstitially.
 4. In dissolving, the hydrogen disrupts slightly the structural networks of the host - causes expansion.
 5. The extent to which this disruption occurs, and the extent of chemical attractive/repulsive forces between the hydrogen and the host, determine the ultimate equilibrium state:
$$\text{H}_2 + \text{M} \rightleftharpoons \text{H}_2 \cdot \text{M} \text{ or } 2 \text{ H} \cdot \text{M}.$$
 6. Solubility is a thermodynamic property, for which enthalpies and free energies are associated.

b. Hydrogen dissolves as the molecular specie. in many materials.

1. Consequently, solubility(S) $\propto$ pressure (H_2).

2. Organic polymers

a) structures generally have atomic-scale voids between twisted polymer chains,

b) H_2 molecules seek those sites.

c)

d) $S \sim 0.1$ cc (STP)/cc.

3. Glasses

a) probably similar to organic polymers around 300 K

b) solubilities $\sim 10^1$ - 10^2 x lower-fewer sites.

4. Ceramics

a) again probably similar to organic polymers around 300 K, but evidence lacking

b) solubilities $\sim 10^{11}$ x lower than polymers (data from extrapolation only).

c. Hydrogen dissolves as atoms in metals and in above materials at higher temperatures.

1. Because $H_2 + M \rightleftharpoons 2H \cdot M$, $S \propto P^{1/2} (H_2)$.

2. Glasses $\geq 300^\circ C$

3. Ceramics (data available $\geq 500^\circ C$)

Hydrogen appears to dissolve and migrate as atoms,
extent of

4. Metals

a) Hydrogen "atoms" sit at octahedral and tetrahedral locations within lattice.

1.

- fcc lattice atoms
- o octahedral sites
- x tetrahedral sites

2. Hydrogen apparently exists within non-hydriding (see below) metal lattices as proton, deuteron, or triton, with electron in metal conduction band.

b) Because some metals are endothermic H_2 absorbers and some are exothermic, there is a great range in solubilities, ~ 10 - 15 orders of magnitude at room temperature.

d. For materials in which there is overall repulsive interaction between hydrogen (whether as molecules, atoms, ions) and constituent molecular framework, solution is endothermic, $\Delta G_S > 0$.

1. Solubility is low, but the higher the temperature, the higher the S.

2. $dS/dT > 0$.

3. True for ceramics, glasses at low temperature (probably), organic polymers, most metals.

e. For materials in which there is overall attraction between the hydrogen specie and the host, solution is exothermic, $\Delta G_S < 0$.

1. Solubility is higher than for endothermic systems, but the higher the temperature the lower S.

2. $dS/dT < 0$.

3. True for glasses at $T \gtrsim 300^\circ\text{C}$ probably because of reduction of Si-O bonds; at 0°C , endothermic?

4. True for some metals because of formation of chemical "hydride" compounds; e.g.,

5. Exothermic hydrogen-absorbing metals are shaded below.

GROUP IA		IIA		IIIA		IVA		VA		VIA			
1	1 00797 H Hydrogen	3	6.931 Li Lithium	5	10.811 B Boron	6	12.01115 C Carbon	7	14.0067 N Nitrogen	8	15.9994 O Oxygen		
11	22.98976928 Na Sodium	13	26.9815386 Al Aluminum	15	30.973761998 P Phosphorus	16	32.065 S Sulfur	17	35.453 Cl Chlorine	18	39.9624 Ar Argon		
19	39.0983 K Potassium	21	44.955912 Sc Scandium	23	47.88 V Vanadium	25	50.9415 Cr Chromium	27	58.933195 Co Cobalt	29	63.546 Cu Copper	31	69.723 Ga Gallium
27	58.933195 Fe Iron	29	63.546 Cu Copper	31	69.723 Ga Gallium	33	74.9216 As Arsenic	35	78.96 Se Selenium	37	85.4678 Rb Rubidium	39	88.90584 Y Yttrium
35	79.904 Br Bromine	37	85.4678 Rb Rubidium	39	88.90584 Y Yttrium	41	92.90638 Nb Niobium	43	98 Tc Technetium	45	102.9055 Rh Rhodium	47	107.8682 Ag Silver
43	98 Tc Technetium	45	102.9055 Rh Rhodium	47	107.8682 Ag Silver	49	114.818 In Indium	51	121.757 Sb Antimony	53	127.60 Te Tellurium	55	127.60 Te Tellurium
51	127.60 Te Tellurium	53	127.60 Te Tellurium	55	127.60 Te Tellurium	57	127.60 Te Tellurium	59	127.60 Te Tellurium	61	127.60 Te Tellurium	63	127.60 Te Tellurium
59	127.60 Te Tellurium	61	127.60 Te Tellurium	63	127.60 Te Tellurium	65	127.60 Te Tellurium	67	127.60 Te Tellurium	69	127.60 Te Tellurium	71	127.60 Te Tellurium
67	127.60 Te Tellurium	69	127.60 Te Tellurium	71	127.60 Te Tellurium	73	127.60 Te Tellurium	75	127.60 Te Tellurium	77	127.60 Te Tellurium	79	127.60 Te Tellurium
75	127.60 Te Tellurium	77	127.60 Te Tellurium	79	127.60 Te Tellurium	81	127.60 Te Tellurium	83	127.60 Te Tellurium	85	127.60 Te Tellurium	87	127.60 Te Tellurium
83	127.60 Te Tellurium	85	127.60 Te Tellurium	87	127.60 Te Tellurium	89	127.60 Te Tellurium	91	127.60 Te Tellurium	93	127.60 Te Tellurium	95	127.60 Te Tellurium
91	127.60 Te Tellurium	93	127.60 Te Tellurium	95	127.60 Te Tellurium	97	127.60 Te Tellurium	99	127.60 Te Tellurium	101	127.60 Te Tellurium	103	127.60 Te Tellurium
99	127.60 Te Tellurium	101	127.60 Te Tellurium	103	127.60 Te Tellurium	105	127.60 Te Tellurium	107	127.60 Te Tellurium	109	127.60 Te Tellurium	111	127.60 Te Tellurium
107	127.60 Te Tellurium	109	127.60 Te Tellurium	111	127.60 Te Tellurium	113	127.60 Te Tellurium	115	127.60 Te Tellurium	117	127.60 Te Tellurium	119	127.60 Te Tellurium
115	127.60 Te Tellurium	117	127.60 Te Tellurium	119	127.60 Te Tellurium	121	127.60 Te Tellurium	123	127.60 Te Tellurium	125	127.60 Te Tellurium	127	127.60 Te Tellurium
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435	127.60 Te Tellurium	437	127.60 Te Tellurium	439	127.60 Te Tellurium	441	127.60 Te Tellurium	443	127.60 Te Tellurium	445	127.60 Te Tellurium	447	127.60 Te Tellurium
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459	127.60 Te Tellurium	461	127.60 Te Tellurium	463	127.60 Te Tellurium	465	127.60 Te Tellurium	467	127.60 Te Tellurium	469	127.60 Te Tellurium	471	127.60 Te Tellurium
467	127.60 Te Tellurium	469	127.60 Te Tellurium	471	127.60 Te Tellurium	473	127.60 Te Tellurium	475	127.60 Te Tellurium	477	127.60 Te Tellurium	479	127.60 Te Tellurium
475	127.60 Te Tellurium	477	127.60 Te Tellurium	479	127.60 Te Tellurium	481	127.60 Te Tellurium	483	127.60 Te Tellurium	485	127.60 Te Tellurium	487	127.60 Te Tellurium
483	127.60 Te Tellurium	485	127.60 Te Tellurium	487	127.60 Te Tellurium	489	127.60 Te Tellurium	491	127.60 Te Tellurium	493	127.60 Te Tellurium	495	127.60 Te Tellurium
491	127.60 Te Tellurium	493	127.60 Te Tellurium	495	127.60 Te Tellurium	497	127.60 Te Tellurium	499	127.60 Te Tellurium	501	127.60 Te Tellurium	503	127.60 Te Tellurium
499	127.60 Te Tellurium	501	127.60 Te Tellurium	503	127.60 Te Tellurium	505	127.60 Te Tellurium	507	127.60 Te Tellurium	509	127.60 Te Tellurium	511	127.60 Te Tellurium
507	127.60 Te Tellurium	509	127.60 Te Tellurium	511	127.60 Te Tellurium	513	127.60 Te Tellurium	515	127.60 Te Tellurium	517	127.60 Te Tellurium	519	127.60 Te Tellurium
515	127.60 Te Tellurium	517	127.60 Te Tellurium	519	127.60 Te Tellurium	52							

58	140.12 Ce Cerium	59	140.907 Pr Praseodymium	60	140.907 Nd Neodymium	61	140.907 Pm Promethium	62	140.907 Sm Samarium	63	140.907 Eu Europium	64	140.907 Gd Gadolinium	65	140.907 Tb Terbium	66	140.907 Dy Dysprosium	67	140.907 Ho Holmium	68	140.907 Er Erbium	69	140.907 Tm Thulium	70	140.907 Yb Ytterbium	71	140.907 Lu Lutetium
90	227.027 Th Thorium	91	227.027 Pa Protactinium	92	227.027 U Uranium	93	227.027 Np Neptunium	94	227.027 Pu Plutonium	95	227.027 Am Americium	96	227.027 Cm Curium	97	227.027 Bk Berkelium	98	227.027 Cf Californium	99	227.027 Es Einsteinium	100	227.027 Fm Fermium	101	227.027 Md Mendelevium	102	227.027 No Nobelium	103	227.027 Lr Lawrencium

54

- f. From the above and from theory of equilibria, for hydrogen-dissociating systems

$$S = S_0 P^{1/2} \exp(-\Delta H_S/RT), \text{ where:}$$

S_0 is a proportionality pre-exponential, a function of entropy

P is pressure

ΔH_S is the enthalpy (heat) of solution, usually in cal/g-atom.

R is gas constant

T is temperature.

- g. Similarly, for equilibria:

$$S = S_0 P \exp(-\Delta H_S/RT), \text{ where } \Delta H_S \text{ is now in cal/g-mole.}$$

- h. From a plot of $\ln S$ versus $1/T$, ΔH_S and S_0 can be extracted:

$$1. \left[\frac{d \ln S}{d(1/T)} \right]_P = -\frac{\Delta H_S}{R}$$

$$2. \ln S_0 = \ln S \text{ intercept at } 1/T = 0$$

3. The following pages show examples of such (Sievert's) plots for various materials.

25

Hydrogen solubility (Sieverts constant) as a function of temperature for selected metals and alloys.

Comparison of Hydrogen Solubility in Refractory Metals to Those in Al_2O_3 , SiC and PyC at One Atmosphere

HYDROGEN SOLUBILITY IN SILICA
AT ONE ATMOSPHERE

- i. Since the solubility of hydrogen in a medium, the driving pressure, and the temperature are mutually related, rearrange

$$S = S_0 P^{1/2} \exp(-\Delta H_S/RT) \text{ to}$$

$$(\ln P)_S = C + 2\Delta H_S/RT$$

1. Differentiating, one obtains

$$\left[\frac{d \ln P}{d(1/T)} \right]_S = 2\Delta H_S/R,$$

another way to determine ΔH_S for a particular composition M_xH_y .

2. An example of such plots is shown for several hydride-forming metals below.

j. The hydride-forming metals exhibit various structural phases, dependent upon temperature and degree of hydrogen loading.

1. For the simple uranium/hydrogen system at some temperature below 650°C :

a) H_2 first dissolves in the metal (α phase) until full homogeneous loading of α has occurred.

b) Upon further loading, hydride phase sites emerge within the lattice and continue to grow until the lattice is ~fully hydride, except for high potential energy α sites.

c) Given enough driving pressure of H_2 , then, uranium goes fully to hydride.

e) $\text{U}(\alpha) \cdot \text{H}$ remains homogeneous to a greater loading the higher the temperature.

f) UH_3 phase remains homogeneous to a smaller loading the higher the temperature.

2. The following phase diagram can thus be written:

3. Pressure-stoichiometry isotherms for the above system are shown below:

- a) Pressure plateaus arise because in $U(\alpha) + UH_3$ section of the diagram there are three phases (including H_2) and two components (H,U). Gibbs' phase rule ($C - P + 2 = F$) indicates one degree of freedom.
- b) Loading and unloading along pressure isotherms is rarely reversible: lattice expansion/compression effects.

4. Similar diagrams for the Zr + H system:

Zirconium-hydrogen phase diagram.

Pressure-composition isotherms in the zirconium-hydrogen system.

k. As noted before, isotope effects on solubility are within factors of 10,

1. See again figure in Section I.E.1.1.2.
2. For the endothermic Ni+H system the following solubility ratios have been found:

Temperature (°C)	Solubility Ratios	
	D/H	T/H
400	0.797	0.709
500	0.805	0.756
600	0.811	0.792
700	0.815	0.823

3. These situations are again primarily functions of zero-point energies.

a)

b) For $H_2(D_2) + Ni \rightleftharpoons 2Ni \cdot H(D)$, $\Delta G_D > \Delta G_H$; D less strongly bound than H; $S(D) < S(H)$.

c) For $H_2(D_2) + U \rightleftharpoons 2U \cdot H(D)$, $-\Delta G_H > -\Delta G_D$; D less strongly bound than H; $S(D) < S(H)$; $P_{S,T}(D) > P_{S,T}(H)$.

2. Please note that the rate of approach to solution equilibrium is a complex kinetic process, being a function of equilibrium physical adsorption (physisorption) of hydrogen gas on a surface:

equilibrium chemical adsorption (chemisorption) of the physisorbed molecule at the surface:

and then the diffusion of the hydrogen specie into the material bulk:

An excellent treatment of a composite process for Ti has recently been published by D. Laser, J. Vac. Sci. Technol., 20, 37 (1982). However, physisorption and chemisorption are generally rapid compared to diffusion. Therefore, of the three only the last process will be discussed.

I.E.2 DIFFUSION OF TRITIUM IN MATERIALS IS IMPORTANT TO CALCULATING THE RATE AT WHICH T WILL MIGRATE THROUGH STRUCTURES. AGAIN WE WILL SPEAK GENERALLY OF "HYDROGEN" AND CONSIDER SMALL ISOTOPIC DIFFERENCES LATER.

a. Migration of hydrogen in most materials is orders of magnitude faster than other elements.

1. Particularly true in metals; for example:

2. However, He often has higher diffusivity than H_2 in polymers, glasses and ceramics.

b. Diffusivity is a kinetic property, a measure of the rapidity with which the diffusing specie jumps from solution site to site.

1. In polymer, for example:

Diffusion is a function of the potential energy barrier between site 1 and site 2.

2. The potential energy contours within the structure may be depicted:

where α is the intersite distance and $\Delta G^\ddagger$ is the activation free-energy for the jump.

3. Since the jump requires activation energy the frequency of jumping is a function of temperature: the higher the temperature the faster the diffusion.
4. Since a loading of sites with solute species would decrease the jumps available to a particular specie, most diffusion determinations are run in the limit of zero solubility.

specie away from its source is a direct function of concentration gradient ($\frac{dc}{dx}$):

$$J = -D \frac{dc}{dx}, \text{ where } D \text{ is the diffusion constant.}$$

1. Consider a crystalline bar with concentration gradient along its axis, and with crystal planes normal to the axis:

2. If there are n_1 diffusing species per unit area in plane 1 and n_2 in plane 2, and if each specie makes Γ jumps/sec, then the total flux of species jumping from plane 1 to plane 2 is:

$$J_{12} = 1/2 n_1 \Gamma.$$

3. Similarly, $J_{21} = 1/2 n_2 \Gamma$.

4. The net flux from plane 1 to plane 2 is then:

$$J = J_{12} - J_{21} = 1/2(n_1 - n_2)\Gamma.$$

5. The concentration of species at plane 1 is

$$c_1 = n_1/\alpha, \text{ and at plane 2, } c_2 = n_2/\alpha.$$

6. So $J = 1/2\alpha(c_1 - c_2)\Gamma$.

7. Since concentration generally changes slowly with distance:

$$c_1 - c_2 = -\alpha \frac{dc}{dx}, \text{ and}$$

$$J = -1/2\alpha^2 \Gamma \frac{dc}{dx}.$$

8. Therefore, $D = 1/2\alpha^2 \Gamma$.

9. In three dimensions, it has been shown that

$$D = 1/6\alpha^2 \Gamma.$$

d. From the above, the temperature dependence of D can be introduced.

1. $D = \gamma\alpha^2 \Gamma$ for general solids, γ relating to geometry of near-neighbor sites.

2. The rate of jumps Γ may be expressed as a function of temperature and activation energy. From rate theory:

$$\Gamma = \Gamma_0 \exp(-\Delta G^\ddagger/RT),$$

where Γ_0 is the frequency of jumps in absence of an energy barrier.

$$\begin{aligned} 3. \text{ So } D &= \gamma \alpha^2 \Gamma_0 \exp(-\Delta G^\ddagger/RT) \\ &= [\gamma \alpha^2 \Gamma_0 \exp(\Delta S^\ddagger/R)] \exp(-\Delta H^\ddagger/RT) \\ &= D_0 \exp(-\Delta H^\ddagger/RT). \end{aligned}$$

4. It has been shown that Γ_0 is on the order of the vibrational frequency of the specie trapped in the diffusion energy well; and hence:

$$\Gamma_0 \propto (1/m)^{1/2}$$

5. If these derivations hold, D_0 should be a function of isotopic mass, $\Delta H^\ddagger$ not explicitly a function.

- e. Consistent with the above development, the temperature dependence of D has been empirically shown:

$$D = D_0 \exp(Q/RT)$$

1. Generally D_0 is $\sim 10^{-3}$ cm²/sec for H₂ in all materials.

2. Q (or $-\Delta H^\ddagger$) varies widely for materials, metals having the lower barriers to diffusion, ceramics the higher; strong, local chemical interaction

interaction M--H is delocalized.

3. Plots for various materials are shown on the following pages.

HYDROGEN DIFFUSION COEFFTS. IN NONMETALLIC SOLIDS

DIFFUSION COEFFICIENTS FOR HYDROGEN IN SILICA

DIFFUSION COEFFICIENTS FOR HYDROGEN IN ALUMINA

CORRELATION OF MELTING POINT WITH
HYDROGEN DIFFUSION ACTIVATION ENERGY

f. In general for most substances, $D(H) > D(D) > D(T)$.

1. In polymers this is not pronounced, see table next page.

2. In glasses and ceramics, isotopic data are sparse, but presumably diffusivities are within a factor of 10.

3. In stainless steels, $D_0 \propto (1/M)^{1/2}$, $Q(D) = Q(T)$, consistent with previous derivations.

Deuterium diffusivity in austenitic stainless steels.

Tritium diffusivity in Type 304L stainless steel.

Handwritten notes:

$1.5 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$3.2 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$5 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$6 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$2.2 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$1.1 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

$1.1 \times 10^{-7} \frac{\text{cc}}{\text{sec}}$

ISOTOPE EFFECTS

$$\frac{\text{cc (STP)} \cdot \text{mm}}{\text{cm}^2 \cdot \text{sec}}$$

$$\text{cm}^2/\text{sec}$$

Membrane Material	Gas	P_{25°	P_H/P_D	P_H/P_T	D_{25°	D_H/D_D	D_H/D_T
Epoxy Resin	H ₂	3.40×10^{-8}	1.12 ± 0.04		2.17×10^{-7}	0.96 ± 0.06	
	D ₂	3.03×10^{-8}			2.25×10^{-7}		
Araldite	H ₂	1.40×10^{-7}		1.17 ± 0.03	3.82×10^{-7}		1.01 ± 0.04
	D ₂	1.20×10^{-7}			3.80×10^{-7}		
Polyethylene 800	H ₂	2.43×10^{-7}	1.12 ± 0.04				
	D ₂	2.18×10^{-7}					
Neoprene	H ₂	0.95×10^{-6}	1.07 ± 0.04	1.09 ± 0.05	2.61×10^{-6}	1.04 ± 0.10	1.07 ± 0.07
	D ₂	0.89×10^{-6}			2.51×10^{-6}		
	T ₂	0.87×10^{-6}			2.43×10^{-6}		
Neoprene	H ₂	1.69×10^{-6}	1.10 ± 0.03				
	D ₂	1.53×10^{-6}					
PVC	H ₂	1.45×10^{-6}	1.16 ± 0.03	1.25 ± 0.05	$\sim 2.5 \times 10^{-6}$		
	D ₂	1.26×10^{-6}					
	T ₂	1.16×10^{-6}					
Latex	H ₂	6.92×10^{-6}	1.11 ± 0.04		$\sim 1.9 \times 10^{-5}$		
	D ₂	6.24×10^{-6}					

4. In pure metals of fcc crystal structure,

$$D_0 \propto (1/M)^{1/2} \text{ and } Q(H) \sim Q(D) \sim Q(T):$$

5. fcc Pd is an exception to this trend, where, extrapolating to high temperatures, tritium would actually diffuse fastest. Probably because of the exothermic nature of Pd/H interaction, data is rather confusing:

Diffusivities are all within a factor of 3, however.

6. For the hydriding bcc metals V, Nb, and Ta, D_0 pre-exponentials are essentially isotope independent. Activation energies, however, scale as $Q(H) < Q(D) < Q(T)$. Most diffusivities are still within a factor of 10 above 0°C.

7. As suggested above, diffusivities in hydride formers are very much a function of hydride stoichiometry:

I.E.3 PERMEABILITY OF HYDROGEN THROUGH MATERIALS IS ULTIMATELY A MEASURE OF HOW RAPIDLY TRITIUM WILL ESCAPE ITS CONTAINMENT.

- a. Permeability is a direct function of both diffusivity and solubility:

$$\phi = D \cdot S.$$

- b. Dimensionally:

$$\phi \left(\frac{\text{cm}^3(\text{H}_2, \text{STP}) \cdot \text{cm}(\text{thickness})}{\text{cm}^2(\text{area}) \cdot \text{sec}} \right) = D \left(\frac{\text{cm}^2}{\text{sec}} \right) \cdot S \left(\frac{\text{cm}^3(\text{H}_2, \text{STP})}{\text{cm}^3} \right)$$

- c. To get equilibrium permeation rate through a substance,

$$P = \phi A / \ell = DSA / \ell, \text{ where}$$

A is the area of substance over which, and ℓ is the thickness of substance through which, permeation occurs. P is generally taken as $\text{cm}^3(\text{H}_2, \text{STP})/\text{sec}$, though sometimes as molecules/sec or moles/sec.

- d. Permeation through a substance does not reach its equilibrium rate immediately, rather it approaches its steady-state value according to the equation

$$\frac{P(t)}{P(t=\infty)} = 1 + 2 \sum_{n=1}^{\infty} (-1)^n [\exp(-Dn^2\pi^2 t/l^2)]$$

From this, one calculates that the time to reach 10% of the equilibrium rate is

$$t_{0.1} = .081 l^2/D,$$

and for 90%:

$$t_{0.9} = .312 l^2/D.$$

So, e.g., through 1 cm thick 304L stainless steel at 300°K,

$$t_{0.1} = .081 \cdot 1^2 / 1.4 \times 10^{-12} = 5.7 \times 10^{10} \text{ sec} = 1808 \text{ yr},$$

while through 304L, 1 mm, 500°K

$$t_{0.1} = .081 \cdot 0.1^2 / 1 \times 10^{-8} = 8.1 \times 10^4 \text{ sec} = 22.5 \text{ hr}.$$

- e. With equilibrium permeability is associated a pressure dependence and an "activation" energy:

$$\begin{aligned}\phi &= D \cdot S = D_0 \exp(-\Delta H_0^\ddagger/RT) \cdot S_0 P^n \exp(-\Delta G_s/RT) \\ &= D_0 S_0 P^n \exp(-(\Delta H^\ddagger + \Delta G_s)/RT) \\ &= \phi_0 P^n \exp(-E/RT).\end{aligned}$$

- f. The following pages display $\log \phi$ versus $(1/T)$ plots for various materials.

1. Note that in general:

a) Hydriding metals, which have high solubilities and high diffusivities, have highest permeabilities.

b) Ceramics, carbides, graphite, etc., which have low S and low D , have lowest ϕ .

2. Some materials have high D but low S and thus have mid-range ϕ . Compare aluminum and stainless steel, for example

$$D(\text{Al}) > D(3\text{xx-stainless})$$

$$S(\text{Al}) \ll S(3\text{xx-stainless})$$

so
$$\phi(\text{Al}) < \phi(3\text{xx-stainless}).$$

Hydrogen permeability as a function of temperature for selected metals and alloys.

Comparison of Hydrogen Permeation Coefficients in Refractory Metals to Those in Al_2O_3 , SiC and PyC at One Atmosphere

HYDROGEN PERMEATION OF SILICA

- g. Isotope effects on ϕ derive from such effects on S, D, and have not in themselves been extensively studied.
1. Since in a particular material
$$S(H) > S(D) > S(T) \text{ and}$$
$$D(H) > D(D) > D(T)$$
usually hold,
$$\phi(H) > \phi(D) > \phi(T)$$
should also hold.
 2. ϕ of the three isotopes should usually lie within a factor of 10.
 3. The next page shows plots of H_2 , D_2 , and T_2 permeabilities in PVC and neoprene. Where the former polymer shows a regular isotopic trend, the latter does not.

PERMEABILITY OF POLYVINYL CHLORIDE TO HYDROGEN ISOTOPES

PERMEABILITY OF NEOPRENE TO HYDROGEN ISOTOPES

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II. COMPATIBILITY OF MATERIALS WITH TRITIUM

J. T. GILL

II.A. COMPATIBILITY OF A TRITIUM/MATERIAL INTERACTION VERY MUCH DEPENDS UPON WHAT PROPERTIES ARE MOST OF CONCERN.

1. MECHANICAL INTEGRITY OF MATERIAL

a. Function of:

1. How well the material dissipates the energy of colliding β -particles.
2. How well the material excludes tritium from its bulk, i.e., solubility, diffusivity.

b. Various Materials:

1. Non-hydriding metals good because electron bands carry energy away from β -track.
2. Ceramics, carbides, etc., good because of low S and D.
3. Silicas intermediate: tritium can penetrate bulk; β disrupts, T_2 reduces Si-O network; embrittlement results.
4. Polymers bad: tritium quickly penetrates material; polymeric networks have little or no energy dissipating properties; network disrupted and reduced; T_2 /polymer interface most disrupted.
5. Hydriding metals particularly bad: simply because brittle salt-like nature of most hydrides.

II.A.2 LOW ESCAPE OF TRITIUM FROM ITS CONTAINMENT (SAFETY AND ENVIRONMENTAL CONCERN).

a. Function of permeability and surface conditions; temperature very important.

b. In order of increasing ϕ : ceramics, graphite < silicates < non-hydrating metals < hydrating metals < polymers.

c. For example:

1. Tritium escape from 304 stainless steel container of 0.3 cm thickness, 1000 cm² surface area, 300K, T_2 at 1 atmosphere:

$$P = \phi A/l = 2.1 \times 10^{-13} \cdot 1000/0.3 = 7 \times 10^{-10} \text{ cc(STP)/sec} \\ = 1.6 \times 10^{-4} \text{ Ci/day } (t_{0.9} = 636 \text{ yr})$$

2. At 800K, however:

$$P = 7.2 \times 10^{-7} \cdot 1000/0.3 = 2.4 \times 10^{-3} \text{ cc(STP)/sec} \\ = 540 \text{ Ci/day } (t_{0.9} = 7.0 \text{ hr})$$

3. While for Al_2O_3 at 800K,

$$P = 4.2 \times 10^{-19} \cdot 1000/0.3 = 1.4 \times 10^{-15} \text{ cc(STP)/sec} \\ = 3.1 \times 10^{-10} \text{ Ci/day } (t_{0.9} = 8.9 \text{ yr})$$

II.A.3 MAINTAINING PURITY OF A TRITIUM SUPPLY WHILE CONTAINED BY A MATERIAL.

a. Source of Impurities

1. T will be isotopically diluted by exchange with available H or D.
2. T will be chemically diluted by formation of methane and water from available organics and some oxides.

b. Various Materials

1. Metals (non-hydriding)

- a) CT_4 from fabrication organics and wash residues
- b) protium from organics and oxide layer hydroxyls
- c) water from iron oxide reduction.

2. Ceramics

- a) protium from exchange with surface and near-surface hydroxyls
- b) CT_4 and protium from any organics present.

3. Glasses

- a) little or no CT_4 if washed
- b) protium from exchange of T with surface and bulk hydroxyl groups
- c) increase in protium impurity a function of T_2 diffusion in glass.

4. Metals (Hydriding)

- a) not usually used in tritium service (structurally) but
- b) if previously loaded with H or D, these will exchange with T.

5. Polymers

Readily (compared to above materials) degraded to form methane, protium impurities.

II.A.4 GETTERING CAPABILITIES

a. Concerns:

1. capacity of getter
2. speed of gettering
3. stability of getter in T_2
4. overpressure vs. temperature characteristic.

b. Materials:

1. Hydriding Metals

- a) good capacity - up to 3 atoms T / atom M.
- b) speed-function of condition, amount of surface - can be high, but subject to oxygen poisoning
- c) generally crumble upon hydriding (H or D or T)
- d) prefer gettering at ~ room temperature, regeneration at accessible temperature
 1. Pd - getter cryogenically, regenerate ~ room temperature
 2. U - getter to $\sim 1 \times 10^{-6}$ Torr at room temperature, regenerate at $\sim 500^\circ\text{C}$
 3. Zr - getter to $\sim 1 \times 10^{-10}$ Torr at room temperature, regenerate at $\sim 800^\circ\text{C}$
- e) properties not affected by β -radiation.

2. Organic Getters - unsaturated alkenes, alkynes
- a) can add one T_2 molecule per organic molecule per unit of unsaturation
 - b) speed - must have catalyst (Pd, Pt, Ir) in close proximity over large surface area; still, takes hours to complete gettering; not oxygen poisoned.
 - c) subject to radiation damage - organic polymerized or degraded
 - d) not regenerable with temperature, may with H_2 flush.

II.B. COMPATIBILITY OF METALS WITH TRITIUM

1. Endothermic Systems

a. Plot of metal permeabilities, shown earlier, displays ϕ as range of values below upper limit.

b. Several factors can influence a metal's ϕ :

1. Surface oxide

a) because the ϕ of H through oxide ceramics is orders of magnitude lower than through metals, a thin surface oxide can markedly reduce effective ϕ through material.

b) If LiD salt is placed in contact with the surface of a permeation specimen, the oxide is reduced, allowing permeation pathway.

c) If a metal undergoes surface oxidation in the presence of hot steam, ϕ will decrease until oxidation is complete.

2. Surface Area of Specimen

a) permeation through the oxide layer appears to be a rate determining step to overall specimen permeation; if the total area of oxide is increased (without increasing cross-sectional area of specimen), effective ϕ will increase.

b) see plot previous page.

3. Cold-working of stainless steels can affect both S and D of hydrogen, but ϕ is generally invariant.

4. Gamma-irradiation of a permeating stainless steel sample does not affect ϕ over several hours.

total dose
≤ 100 rad

5. T₂ β-irradiation of the surface of a permeating metal could, however, introduce defects into the surface oxide barrier layer over months (via self-catalyzed reduction) and thereby increase overall ϕ .

- c. Embrittlement of non-hydriding metals can be caused by tritium at high pressures.
1. Tritium diffuses into, solubilizes in metals in significant concentrations when driven at high pressure; concentrations of several hundred or thousand atom ppm are attainable at 5 to 25 kpsi.
 2. Some loss in ductility of metal seen from hydrogen charging alone: 20% loss in elongation of 304L stainless steel when charged at 10 kpsi (classical hydrogen embrittlement).
 3. Tritium charged metals, aged for several years, show additional loss of ductility: 20% loss in elongation (5.5 years after 304L charge with 10 kpsi T_2) compared to unaged specimen.
 4. Caused by He^3 buildup in metal lattice; He^3 mobility in the lattice is orders of magnitude less than T mobility, cannot escape.
 5. Helium embrittlement appears at ≥ 300 atom ppm He; He^3 three to four times more effective than H (on an atom ppm basis) in causing mechanical degradation.
 6. Helium collects at microinclusions or grain boundaries and exerts internal pressure or decohesion forces.

d. Contamination of or by the oxide layer on a metal can be of concern.

1. Contamination of tritium with protium or water derived from the oxide has been discussed - most hydroxyls in oxide will exchange with tritium, given a few torr-hours' exposure.

2. Conversely, contamination of the oxide layer by tritium can be a problem, especially when servicing a metallic item.

a) Most exchangeable (and therefore biologically hazardous) tritium present in a material previously subjected to tritium exposure (e.g., 1 atm, 1 yr), is present in the surface oxide.

b) This tritium is present as hydroxyls (tritoxyls); since ~50% of all oxygen atoms in an oxide layer are bound to a hydrogen atom, a lot of tritium can be present on a rough metallic surface.

c) Removal of tritium from a contaminated metal surface is not easy. Some methods:

1) flush with moist air initially - H_2O will exchange with $-M-OT$,

2) bathe in hot steam, evacuate steam alternately,

3) wash with water/acid solution repeatedly to dissolve away oxide,

4) bombard surface with hydrogen atoms and molecules produced in a glow discharge (proposed).

d) All of the washing methods above have the disadvantages of being time consuming and producing radioactive waste materials - bath solutions, gases, gloves, rags, etc. It is often more cost effective to discard and replace a malfunctioning item than to decontaminate it to facilitate repair.

II.B.2 EXOTHERMIC SYSTEMS

- a. Again, ϕ quite high for most of these metals, even for many alloys incorporating them: should be used with caution in constructing primary tritium containment systems.
- b. These materials are used as hydrogen pumps (getters); pressure vs. stoichiometry isotherms have been shown for U and Zr, two of the more common getters in vacuum systems.
 1. From the $\log P$ vs. $1/T$ plots for plateau region UH_X also shown before, note that overpressures decrease in the series $UT_X > UD_X > UH_X$, for a given temperature.

a) Equations for these plots have been determined:

$$UT_X \log P_{\text{torr}} = \frac{-4471}{T} + 9.461$$

$$UD_X = \frac{-4500}{T} + 9.40$$

$$UH_X = \frac{-4450}{T} + 9.20$$

b) It is therefore possible to purify tritium gas isotopically by sorption and evolution of T_2 in several getter stages.

- c) If T_2 gas goes on U at a certain isotopic purity, it will come off at a higher purity.
2. A more detailed version of Zr/H pressure vs. stoichiometry isotherms is shown below.

Curves for a Zr-Al getter alloy (SAES Co. #ST101) have no plateau regions, indicative of only one phase above 0°C. Lattice expansion, but no rearrangement of lattice metal atoms, accompanies hydrogen sorption.

c. Upon formation of hydride phase in the metal phase, mechanical properties are severely affected.

1. Lattice constants of hydride inclusions are increased, densities decrease:

U	19.07 g/cm ³	UT ₃	11.29
UH ₃	10.91		
UD ₃	11.10		

2. Ultimate tensile strength of specimen decreases, ductility increases until pure (brittle) hydride phase is formed, where ductility decreases precipitously.

Mechanical usefulness of Zr metal is limited above ~0.4 H/Zr ratio from 0-300°C.

- d. Oxidation of getters is also an exothermic process - and the oxygen is not regenerable.
1. Oxide layers on the surface of getters prevent sorption of hydrogen just as such layers decrease permeation rates through endothermic metals.
 2. Surfaces of getters must initially be activated to disrupt the oxide barrier layer.
 - a) High temperatures ($>500^{\circ}\text{C}$) will drive oxygen from layer (by diffusion) into bulk.
 - b) Slightly lower ($>250^{\circ}\text{C}$) will allow sufficient penetration of H through layer to affect metal structure and undermine layer.
 3. Examples
 - a) Hydriding of massive U metal may be initiated by heating briefly to 300°C in the presence of hydrogen.
 - b) Significant hydrogen pumping speed onto Zr alloy panels is obtained by heating for several minutes to $500-900^{\circ}\text{C}$, depending on the alloy and the pumping speed desired.

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II.C. COMPATIBILITY OF TRITIUM WITH CERAMICS/GRAPHITE/GLASSES.

1. Interactions between tritium and these materials (at room temperature at least) are generally limited to surface and near-surface regions; S, D, ϕ are quite low.

2. Graphite

a. Because of generally high surface areas, graphite samples adsorb large amounts of hydrogen gas: 4×10^{18} molecules/gram for a graphite pellet used in gas-cooled reactors.

b. Methane, protium, and probably some water are generated from the β -irradiation of the graphite surface.

c. T_2 gas can be contaminated by several percent impurities after weeks-months, depending on T_2/C ratios.

- d. Surface of graphite will be contaminated with chemically-bound T; decontamination possible through baking at $\geq 500^{\circ}\text{C}$ in presence of a hydrogen exchange medium (H_2 , H_2O , NH_3 , etc.).
- e. Graphite will probably not be mechanically degraded over several years.

3. Glasses

- a. Permeability of silicas and borosilicate to D_2 are anomalously low at $\geq 300^\circ C$ when measured over short times:

- b. The above permeabilities recover to a straight plot, however, after longer times.
- c. Following the permeation measurements, solubility of deuterium was determined by outgassing and subsequent flushing by H_2 - far more deuterium could be flushed than outgassed.

- d. Partial reduction of glasses in hydrogen at temperatures $>300^{\circ}\text{C}$ has thus been proposed; infrared data show increase of Si-OH band, presumably from:

- e. Other studies show that silicas, soaked in D_2 at 200°C and then subjected to γ -irradiation, show Raman bands of Si-D and Si-OD species, as well as dissolved D_2 :

- f. The above data suggest that T_2 gas in the presence of its chemically activating β -irradiation energy could reduce silica bonding to $-Si-T$ and $-Si-OT$ species.
- g. Migration of tritium into glass structures could thus cause embrittlement and potential fracture over several months/years - embrittlement of borosilicate in presence of tritiated water experienced by author:

- h. Evidence to suggest crystallization of glassy silica structures under high energy electron bombardment also exists.
- i. Exchange of tritium with hydroxyl groups naturally occurring in various glasses (and on their surfaces) is a source of protium contamination to tritium - perhaps 1% HT into 1 % T_2 gas at 1 atm. after 1 year.
- j. Decontaminating a well-soaked glass item of its bound tritium would require significant investment in H_2O washes or higher temperature H_2 permeation flushes.

4. Ceramics

- a. Situation somewhat similar to glasses; e.g., alumina bonds could also be reduced in tritium.
- b. However, S , D , and ϕ are generally so much lower for ceramics than for glasses, that the former would, comparatively, undergo little or no bulk disruption.
- c. Some reduction and mechanical degradation of near-surface regions possible, depth of affected area being a function of D and time.
- d. Exchange of surface and near-surface protium is likely, though mutual contamination of T_2 and material should again be lower than glass; study on Al_2O_3/T_2 showed little or no protium in gas.
- e. Decontamination of a tritium-contaminated item possible via water, acids, and heat; still probably not cost effective.

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II.D. PLASTICS/ELASTOMERS/OILS

1. Organics are readily penetrated by tritium and are thus subject to disruption of their chemistry in bulk.
 - a. There are few or no mechanisms for rapid delocalization of π -energy; aromatic systems provide the greatest dissipation, but even these are limited.
 - b. There is substantial mobility of organic chains within polymer structures; reactive organic intermediates, once formed, can react with each other.
2. Several mechanisms of polymer chain modification occur following activation by β -radiation to ionic or excited species.

a. Initially:

i) Main-chain rupture

ii) Side-chain rupture

b. Followed by:

i) Cross-linking: the formation of chemical bonds between two polymeric chains

ii) Unsaturation: the formation of double chemical bonds within the polymer chain

iii) Degradation: the fracturing of polymer molecules

iv) Gas evolution, which results in the rupture of small molecules from the polymer

c. T_2 can then react with any of the above materials, inhibiting cross-linking and unsaturation, promoting degradation.

3. Cross-linking and degradation are the processes most important to the mechanical properties of the polymer.

a. Both processes compete in a material, but those polymers which are most sterically hindered appear to preferentially degrade.

b. Of the vinyl-type polymers,

degrade and

cross-link.

c. Steric hindrance prevents neighboring chains from linking also imparts structural strains which are relieved upon chain scissioning.

d. Cross-linking is noted mechanically by an increase in tensile strength but decrease in elongation to break: embrittlement.

e. Degradation is evidence by a softening toward a gummy consistency: increase in elongation, decrease in tensile strength.

4. Several factors that affect relative polymer stability have been determined in the γ literature.
- a. Energy-delocalizing aromatic structural groups increase polymer stability by distributing energies of excited states.
 - b. Halogen atoms within polymers are efficient at generating free radicals and thus radiation damage.
 - c. Substituents on aromatic groups which extend the delocalized bonding network are further stabilizers.
 - d. Saturated aliphatics are more radiation-resistant than unsaturated ones; isolated double bonds are readily excited to ions or radicals.
 - e. Organic compounds, in order of decreasing radiation resistance are: aromatics, aliphatics, alcohols, amines, esters, ketones, acids.
5. The following pages show the relative radiation resistances of plastics, thermosets, elastomers, and oils. The relationships generally follow the above guidelines and are probably applicable in large part to T_2 β -irradiation.

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RELATIVE RADIATION RESISTANCE OF THERMOPLASTIC RESINS

A-46921

RELATIVE RADIATION STABILITY OF THERMOSETTING RESINS

A-46920

RELATIVE RADIATION STABILITY OF ELASTOMERS

Radiation resistance of base oils

II.D.6. The dose in rads imparted to a material by exposure to tritium β -radiation can be estimated.

a. Bulk Material

1. Function of T_2 solubility, time.
2. For teflon of density = 2.2
dissolving 0.04 cc (T_2 , STP)/cc:

$$\text{Dose} = \frac{0.04 \text{ cc } T_2}{\text{cc}} \cdot \frac{\text{cc}}{2.2 \text{ g}} \cdot \frac{2.6 \text{ Ci}}{\text{cc } T_2} \cdot \frac{3.7 \times 10^{10} \text{ dis/sec}}{\text{Ci}} \cdot \frac{5.7 \times 10^3 \text{ eV}}{\text{dis}} \cdot \frac{1.6 \times 10^{-12} \text{ erg}}{\text{eV}} \cdot \frac{\text{rad}}{10^2 \text{ erg/g}} \cdot$$

$$\frac{3.2 \times 10^7 \text{ sec}}{\text{yr}} = 5 \times 10^6 \text{ rad/year.}$$

3. For teflon, one year's exposure to T_2 at STP would cause severe bulk damage (as suggested by γ -literature).

b. Surface of Material

1. Function of range of β -particle in T_2 , range in material, height of T_2 column above surface

2. For a planar teflon surface with several cm height of T_2 at STP above it:

- a) The literature gives the energy incident on the solid surface from the gas phase as:

$$E_i = E_o / 4 \mu$$

where E_o = energy release per sec per cc of gas

μ = absorption coefficient of gas

- b) Since $\mu(T_2) \approx \mu(H_2)$, for the teflon example:

$$\begin{aligned} E_i &= E_o / 4 \mu = \frac{3.7 \times 10^{10} \text{ dis} \cdot \frac{2.6 \text{ Ci}}{\text{cm}^3}}{\text{sec} \cdot \text{Ci}} \\ &= \frac{5.7 \times 10^3 \text{ eV}}{\text{dis}} / 4 \cdot 1.81 \text{ cm}^{-1} \\ &= 7.6 \times 10^{13} \text{ eV/sec} \cdot \text{cm}^2 \end{aligned}$$

- c) Converting to rads:

$$\begin{aligned} E_i &= 7.6 \times 10^{13} \text{ eV/sec} \cdot \text{cm}^2 \cdot 1.6 \times 10^{-14} \\ &= \text{rad} \cdot \text{g/eV} / 2.2 \text{ g/cm}^3 \\ &= 0.55 \text{ rad cm/sec.} \end{aligned}$$

Handwritten calculations:
 $1.6 \times 10^{-14} \text{ rad}$
 $7.6 \times 10^{13} \text{ eV/sec} \cdot \text{cm}^2$
 $1.6 \times 10^{-14} \times 7.6 \times 10^{13} = 1.216$
 $1.216 / 2.2 = 0.5527$

d) And since the range of the β in teflon is short,

$$\sim 2.3 \times 10^{-5} \text{ cm},$$

$$\text{DOSE} \approx 0.55 \frac{\text{rad.cm}}{\text{sec}} \cdot \frac{1}{2.3 \times 10^{-5} \text{ cm}} \cdot 3.2 \times 10^7 \frac{\text{sec}}{\text{yr}}$$

$$= 7.7 \times 10^{11} \text{ rad/yr.}$$

3. Roughly, then, the top layer of surface receives $\sim 10^5$ x the dose that the bulk experiences. After one year, the surface of teflon would probably be chemically vaporized to CT_4 and TF. Other plastics of greater resistance might be surface embrittled or degraded ("tacky").

7. Some direct experience of polymers with tritium has been obtained.

a. Teflon or Kel-F exposure in T_2 does indeed produce TF.

1. Noted as SiF_4 gas in glass system.

2. Because of this acid production, T_2 + moisture + teflon in a stainless steel system at pressures ~ 1300 atm caused catastrophic stress corrosion cracking of 0.76 mm-thick steel in 16 hours; substituting D_2 for T_2 , or removing teflon or moisture caused no failure.

Catastrophically failed specimen.

- b. The relative increase in hardness of several elastomers after 87 days exposure in 100 Torr T_2 was investigated.
 1. Hardening increased in the following order: natural ~butadiene/styrene~chlorosulfonates < butadiene/acrylonitrile~chloroprene < silicone (crumbled).
 2. This series follows quite well the relative stabilities as determined under γ -radiation.
- c. Total compression set at 100 atm•days T_2 exposure: _____
 EPR < SBR < Vitons < nitrile < urethane < silicone.
- d. Surface vs. bulk effects have been noted in polymer/ T_2 studies.
 1. Hardening of neoprene (above) occurred throughout the bulk, while that of natural rubber primarily occurred at the surface (crack propagation).

2. Total incorporation of tritium into a polyethylene powder was not a function of the amount of powder but of the exposed surface area.

Explanation: greater concentration of reactive T₂ atoms (and greater radiation field) at T₂/polymer interface than in bulk.

3. Radiation-induced fluorescence from the surface of T₂-exposed HDPE orders of magnitude greater than that from the bulk.

8. Some other tid-bits:

- a. Polyimides (good in γ service) appear good in T₂ service.
 1. Plastic stem tips for valves when used with sufficient sealing force, continue to seal for several years in T₂ (~STP).
 2. When employed with less sealing force, however, leaks are noted across valve tip; possibly because of surface hardening.
 3. Polyimide gaskets, under constant sealing load, probably adequate for years.

b. After 130 days exposure in T_2 at ~ 1.5 atm, increase in polymer hardness fell in the series:
Vespel (polyimide) $\sim$ Kalrez (perfluoro) $<$ Viton (perfluoro)

c. Pump Oils

1. Saturated hydrocarbon mineral oils (e.g., Duo-Seal) require frequent changes in T_2 service
 - a) vapor pressure increases - offgassing
 - b) liquid viscosity increases markedly.
2. Polyphenyl ether oils last for years in the same service, but are very expensive

3. Perfluorinated pump oils not recommended for T_2 service: probable TF evolution.

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II.E. ELECTRONIC EQUIPMENT

1. β -radiation can probably disrupt semiconductor operation at low dose ($\sim 10^5$ rad), as does γ .
2. Because of ions and electrons present, T_2 gas can act as a "leaky dielectric".
 - a. Measurement of T_2 pressure via direct-contact capacitance manometry is often grossly in error.

ZERO SHIFT AS A FUNCTION OF PRESSURE FOR A MKS 221A 1000 TORR DIFFERENTIAL CAPACITANCE MANOMETER WHEN BOTH P_X and P_R SIDES ARE EXPOSED TO EQUAL PRESSURES OF TRITIUM.

- b. Applying a voltage bias to a plate in T_2 gas will allow leakage of current to ground and cause gaseous ("glow") discharge conditions:

3. Electron multipliers or scintillation detectors used in a tritium-contaminated environment will experience a slow, irreversible increase in background counting rate, due to tritium absorbed in the surfaces of the materials of construction; β 's from this residual tritium will be counted.

Background signal from a Galileo 4501 CEM as a function of cumulative T_2 exposure quantity (in Pa-s).

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TRITIUM HANDLING
IN VACUUM SYSTEMS
PART TWO

A Short Course Offered by
The American Vacuum Society
Baltimore, Maryland

16 November 1982

Instructors:

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A. General Considerations - Materials

To produce useful electrical energy continuously, a fusion plant will require a tritium inventory of many kilograms (Fig. 1). All of this material must pass through the plasma chamber evacuation system many times per day enroute to the fuel processing facility, so the vacuum engineer is confronted with the problem of handling large quantities of this radioactive fuel. Many processes in the overall fuel purification loop also require subatmospheric handling of tritium-laden gas mixtures under a wide range of conditions (Fig. 2).

At first the problems of containing tritium in a metal system seem no worse than those of containing ordinary hydrogen. Structural components must not be permeable to hydrogen nor subject to hydrogen embrittlement in the temperature and

Fig. 1. Tritium inventories at fusion facilities.

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Fig. 2. Tritium parameters for fusion fuel processing.

pressure regime where they are used (Fig. 3). The toxic, radioactive, expensive nature of tritium only makes these requirements more stringent. Austenitic stainless steels provide adequate strength and integrity for vacuum vessels at ambient and cryogenic temperatures. For processing tritium at temperatures above 600 K, stainless steels are not satisfactory because of the high permeability of iron/nickel alloys to hydrogen. Copper and aluminum have much lower permeabilities, but they also have reduced strength and creep resistance at high temperature. One solution is to use a composite vessel with a liner of low permeability reinforced by a high-strength outer shell. For temperatures above 800 K ceramic vacuum vessels must be considered. Alumina has very low permeability to

Fig. 3. Tritium containment problems.

hydrogen at high temperatures, but it is an extremely difficult material to fabricate into complex shapes.

A special problem of containing tritium, not experienced with hydrogen or deuterium, involves the choice of soft sealing materials--compression gaskets, valve packings and seats, etc. The average 6-keV Beta particle carries 1000 times more energy than is required to break the strongest organic bond. Tritium, like hydrogen, forms compounds with nearly every element in the periodic table, it exchanges with ordinary hydrogen in organic compounds, and the energy available from its radioactivity catalyzes reactions at ambient conditions that would normally occur only at elevated temperatures. For example, tritium gas stored at ambient temperature in stainless steel attacks the carbon exposed at the grain boundaries and

synthesizes tritiated methane spontaneously (Fig. 4). Such reactions accelerate the decomposition of all organic materials. In fact the useful life of most elastomers chronically exposed to tritium is measured in days or even hours. Some organic materials also release highly corrosive decomposition products when attacked by tritium, a fact that can have serious consequences if overlooked. Fluorocarbons, which release tritium fluoride as they decompose (Fig. 5), are particularly nasty examples. An epidemic of metal bellows failures was once traced to indiscriminate use of fluorocarbon valve seats and gaskets in a tritium system (Ref. R-5).

Fig. 4. Spontaneous chemical synthesis by tritium.

Fig. 5. Decomposition of plastics by tritium exchange.

The solution is to use metal gaskets wherever possible and to design any necessary plastic seals so that only the minimum area surface is exposed to the tritium vacuum medium. Fortunately, bakeable high vacuum systems have the same requirements (freedom from organic materials) and for that reason they are usually also tritium-compatible. Where the resilience or low friction of a plastic is absolutely required, as in soft-seat valves, the polyimide or polyimide-amide resins have proven to be more radiation resistant than most. These plastics are also good high temperature materials, and they are used as seat material in bakeable high vacuum valves.

B. Piping and Fittings

The same materials discussed above for construction of vacuum vessels and components usually are favored for tubing and ducting in vacuum systems that will handle tritium. The most common are types 304L and 316L stainless steel, deoxidized copper (DPE or DHP), and aluminum alloys, with the stainless steel components dominating the inventories of most vacuum equipment houses. Advantages of copper that should not be overlooked are its lower permeability to hydrogen, availability of high purity (particularly oxygen-free) forms, and the ease of fabrication with fully annealed copper tubing. Aluminum offers many of the same advantages and in addition it has a very low activation cross-section for neutrons, an important consideration for components located near a fusion reactor. It is more difficult to weld aluminum reliably, and joining it to components made of other materials is a problem, though stainless-to-aluminum transitions are now available in pipe diameters up to 800 mm (Ref. M-28).

Metal-sealed high-vacuum fittings are commercially available in sizes from 3 mm to over 1 meter. At the small end of the scale are the zero-axial-clearance fittings (Ref. M-4) with their throw-away metal compression gaskets for tubing sizes to 25 mm. Since these fittings seal with a butt-type gasket, components mounted with them can be removed without disturbing the connecting tubing, a very desirable feature for glove box installations. Next in size are the copper-sealed knife edge flanges (Fig. 7). These are normally available off

VCR Cross

VCR Double Female Union

VCR Double Male Union

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Fig. 6. Metal-gasketed zero-clearance tube fitting
(Courtesy of Cajon Co., Macedonia, OH).

CUSEAL ROTATABLE FLANGE

Fig. 7. Vacuum flange with copper knife-edge gasket
(Courtesy of Granville-Phillips Co., Boulder, CO).

the shelf for tubing sizes up to 200 mm. In the intermediate sizes also, the European standard vacuum flanges of the ISO-KF (10 to 50 mm) and ISO-LF (62 to 1000 mm) types are becoming popular in this country. All come with well designed assembly bolt-up systems, and various types of all-metal sealing systems are available, though the original flanges were designed for elastomer O-rings (Fig. 8). For vacuum ducts of 500 mm and larger, a variety of wire-sealed (Fig. 9) or aluminum-foil-sealed flanges are available (Refs. M-12, M-15, M-20, M-24, M-25, M-26). Figs. 10 and 11 show some of the metal seals intended to seal flat or grooved flanges.

Fig. 8. ISO small flange system with metal seals
(Courtesy of Alcatel Vacuum Products, Hingham, MA).

Fig. 9. Wire-ring seal system for large flanges
(Courtesy of Varian Vacuum Div., Palo Alto, CA).

Fig. 10. Machined and formed all-metal flange seals
(Courtesy of Advanced Products Co., North Haven, CT).

Fig. 11. Spring-loaded all-metal seals for large flanges
(Courtesy of Helicoflex Co., Boonton, NJ).

C. Vacuum Valves

Tritium-compatible valves are difficult to fabricate for three reasons. First, the sealing action must be made repeatedly and reliably with tritium-compatible materials at the valve seat. Second, the sealing motion must be introduced through the vacuum wall hermetically. Third, most valves on tritium-handling systems are of necessity located inside glove-boxes, so the valve must be operated by remote control. The second problem was solved long ago by the use of metal-bellows-sealed stem actuators for both linear and rotary motion. Vacuum-grade valves of stainless steel or aluminum with bellows-sealed stems are available from many manufacturers. Most of these valves are available with pneumatic operators, so the

TRITIUM-COMPATIBLE VACUUM EQUIPMENT

third requirement above is easily satisfied. Valve bonnets may either be welded or sealed with one of the metal gaskets discussed in the previous section. Most general service vacuum valves come assembled with elastomeric O-rings, so the tritium designer must ensure that all such seals are replaced with tritium-compatible gaskets.

For small lines carrying tritium gas mixtures at viscous flow conditions, a variety of valves are already in use in the nuclear (fission) industry. One manufacturer's line of valves (Ref. M-19) features bellows-sealed stems, 316 SS bodies, metal or polyimide-tipped valve stems, and the bellows either welded to the valve body or sealed with metal O-rings for quick replacement (Fig. 12). Sizes run from 3 to 20 mm tubing,

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Fig. 12. Bellows-sealed and stem-packed valves compared
(Courtesy of Nupro Co., Willoughby, OH).

and a complete line of pneumatic actuators for remote operation is available.

In the more usual tube sizes for medium and high vacuum work (20-100 mm), a few metal- or polyimide-sealed valves are available. Several vacuum component manufacturers make polyimide-sealed valves (Fig. 13) in 15- to 38-mm sizes (Refs. M-25, M-26). The traditional gold-seat bakeable ultra-high vacuum valve (Fig. 14) is another possibility, but these do not adapt readily to remote operation because of the high seating loads required. A very reliable all-metal high vacuum valve has been developed in Europe (Ref. M-39) and is now marketed in the United States (Ref. M-11). The valve (Fig. 15) uses a hardened conical nosepiece sealing against a Belleville

Fig. 13. Pneumatic-operated, polyimide-seat vacuum valve (Courtesy of Varian Vacuum Div., Palo Alto, CA).

Fig. 14. Bakeable gold-seat ultra-high-vacuum valve
(Courtesy of Granville-Phillips, Boulder, CO).

Fig. 15. Metal-sealed valve with Belleville washer seat
(Courtesy of HPS Corporation, Boulder, CO).

washer seat, a design that minimizes permanent inelastic deformation. These valves are available in right-angle designs for pipe diameters up to 150 mm, and in pendulum-gate models up to 630 mm, both types in pneumatic and manual versions.

Variable leak valves have many applications in tritium treatment systems, and several types already on the market for other purposes are intrinsically compatible with tritium. One type of variable leak valve (Ref. M-26) exposes only metals and a sapphire seat to the vacuum medium (Fig. 16). Another model of all-metal leak-control valve (Ref. M-9) can be servo-motor-controlled by any process parameter convertible to a d.c. voltage.

Fig. 16. Sapphire-seat variable-leak high vacuum valve
(Courtesy of Varian Vacuum Division, Palo Alto, CA).

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The final item under the subject of valves is the pressure relief device. Tritium-compatible relief valves that seal reliably when not relieving are quite difficult to make. Because the relief valve must be vacuum-tight below its relief pressure, it usually relies upon an elastomer-sealed seat. Since the elastomer is chronically exposed to the tritium in the vacuum medium, it soon loses its ability to seal. For absolute relief of unexpected overpressures metal rupture disks are therefore usually the best choice. Of course for normally evacuated components the disk should either be of the reerze-buckling type or be installed with a vacuum support. Aluminum, which is tritium-compatible, is the disk material most used for low pressure relief. If there is any chance that tritium might be vented when the disk ruptures, the exhaust must be ducted to a holding tank for subsequent recovery or for tritium removal. If the holding tank is evacuated, the designer should remember that the pressure rating of the relief device refers to the differential pressure across the disk.

Where a small leakage across the relief valve seat can be tolerated, and where relief action is expected to occur intermittently, the sliding-gate back-pressure regulating valve is a very satisfactory device. Such an application might be regulation of the maximum pressure on the discharge of a metal-bellows pump by relief of the surplus pressure back to the pump inlet. This type of valve (Fig. 17) regulates pressure by sliding a lapped metal plate over an orifice under the control of a spring-loaded metal diaphragm. One manufacturer

Fig. 17. Sliding-gate pressure-regulating valve
(Courtesy of Jordan Valve, Cincinnati, OH).

offers these units in on/off, pressure-reducing, differential, and vacuum range models, as well as the back-pressure regulator (Ref. M-30). They are available in spring- and pneumatically-loaded, and motor-actuated versions in a wide variety of pressure ranges, pipe sizes, and flow ratings. To "harden" the factory supplied valves for tritium service, it is necessary to disassemble and degrease the sliding disk and seat. On re-assembly the existing gasketless body seals should be supplemented by groove-confined wire-ring seals, and suitable fittings for tritium should be brazed or welded to the inlet and outlet.

Although most remote-operated vacuum valves for tritium systems will reside in glove boxes (see Secondary Containment, Sec. F), this may not be practical for very large vacuum gate valves in which the tritium is always at very low densities (high vacuum). Where no overall secondary containment is used, the valve-stem bellows should always be secondarily enclosed and means provided for detection of leaking tritium. Fig. 18 illustrates a method that accomplishes this. Argon or other inert gas can be circulated through the bonnet of such a modified valve and then through an ion chamber to detect any tritium that might have leaked from a failed bellows.

Fig. 18. Secondary containment and leak detection of bellows-sealed valves.

D. Vacuum Gauges

With few exceptions, standard vacuum gauges will work for tritium without any special modifications. Thermal conductivity gauges (thermocouple and Pirani) must be calibrated for each specific mixture of hydrogen isotopes, and they generally have poor calibrations for the hydrogens. Gauges that are insensitive to any properties of a gas except its pressure are of course preferred. Mercury manometers and McLeod gauges are satisfactory gauges for tritium, but designers may prefer to avoid mercury for other reasons (see comments under Vacuum Pumps, Sec. E). Mechanical gauges (Bourdon, aneroid) are suitable for tritium, but the capsule material should be chosen to minimize tritium permeation if the gauge element is to be exposed to tritium at high temperatures.

In recent years significant advances have been made in capacitance manometers, particularly in one-side designs, in which the measured capacitance is on the permanently evacuated or reference side of the gauge (Fig. 19). For absolute pressure measurements between 1 atmosphere and 0.1 millitorr the single sided capacitance manometer is probably the best general purpose vacuum gauge available today for use in tritium-vacuum systems. The capacitance manometer is not a good choice for differential pressures however, because ionization from tritium on the reference side tends to short out the capacitance, and large measurement errors result. This is true for double sided capacitance manometers as well. A variable reluctance (LVDT) diaphragm or capsule transducer (Ref. M-40) may

TRITIUM-COMPATIBLE VACUUM EQUIPMENT

Schematic of single-sided sensor.

Welded assembly has sealed reference side.

Fig. 19. Single-sided capacitance manometer (Courtesy of MKS Instruments, Burlington, MA).

Fig. 20. Variable reluctance (LVDT) differential pressure transducer (Courtesy of Shaeffert Engineering, Pennsauken, NJ).

be a better solution for tritium differential pressure measurements because the electronics are not exposed to the vacuum medium on either side of the diaphragm (Fig. 17). Of course such a gauge must employ a metal membrane pressure element, and all static case seals must be tritium-compatible.

For the high vacuum range, ion gauges of either the hot filament or cold cathode type work as well for tritium as for other isotopes of hydrogen. Any ion gauge must be calibrated for the particular gas with which it will be used, and the error from tritium ionization is insignificant at the pressures at which ion gauges operate. Of course gauges with glass envelopes should not be used in tritium systems because of the risk of breakage. Metal envelopes, or nude gauges mounted entirely within the vacuum enclosure, should be utilized instead. Where isotopic partial pressures are of interest, the quadrupole mass spectrometer is an especially useful adaptation of the basic ion gauge.

E. Vacuum Pumps for Tritium

Vacuum pumps for tritium service must have the same features as the other tritium-compatible components already discussed. The main requirement is that absolutely no organic lubricants, elastomers, or fluorocarbons be used in the process-wetted passages of the pump. Since most mechanical pumps require continuous rotary or reciprocating motion, tritium compatibility is a severe limitation on their selection and design. For this reason conventional oil-sealed reciprocating-piston or rotary-vane pumps will not be discussed further, except under house vacuum systems (Sec. E.4).

One class of pumps that have been used successfully for tritium are those that use mercury or mercury vapor as the pumping fluid. Mercury pumps working on many different principles can be built for the rough vacuum (Toepler and Sprengel), vacuum booster (mercury injector), or very high vacuum (mercury diffusion) regimes. Although mercury is completely compatible with tritium, it amalgamates with many metals commonly used in vacuum systems. It is so mobile, even in the liquid state, that it is virtually impossible to confine it to the devices in which it is used. Because mercury attacks so many useful vacuum materials (silver, copper, aluminum, etc.), the designer of fusion-related vacuum systems will generally want to avoid it entirely. Fortunately, there are now several other choices to replace mercury pumps in all the vacuum regimes. For those readers wishing more information these mercury pumps

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are described in reprint R-4, included as an appendix to these notes by permission of the authors.

Most tritium pumping problems can be solved either with a mechanical pump with special hermetically sealed drives (Fig. 21), or by a class of pumps that have no moving parts, called reservoir or "capture" pumps. Table I lists a number of vacuum pumps that have been considered for tritium, their pressure ranges, compression ratios, and pumping speed ranges. The conventional oil-sealed rotary pump is listed for comparison only --it is not a suitable tritium pump under any circumstances.

E.1. Roughing Pumps

Oil-sealed rotary and mercury gas transfer pumps have already been discussed. The next type of pump the designer is likely to consider for tritium is the "oil-less" mechanical vacuum pump. Three common types are sold under this general label: 1) "dry" rotary vane, 2) oil-less piston, and 3) diaphragm. These pumps are sold principally for rough vacuum OEM applications, and none of them will produce particularly good vacuums. Of course the vacuum attained may be adequate for a specific application, but all these pumps have other drawbacks for tritium service. First, the shaft packings or piston seals in the first two types are not necessarily vacuum-tight. This may not be important for the applications for which these pumps are designed, but it does preclude their use with tritium. Furthermore, their "oil-less" packings are most often fabricated from self-lubricating fluorocarbons, which are not

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**BELLOWS
TRANSFER
PUMPS**

**MAGNETIC-BEARING
TURBOMOLECULAR
PUMPS**

MAGNETIC DRIVES, ETC.

Fig. 21. Special constructions for tritium pumps.

TABLE I. CHARACTERISTICS OF TRITIUM-COMPATIBLE VACUUM PUMPS

Pump Type	P_{min} (Pa, abs)	P_{max} (kPa, abs)	P_{out}/P_{in} (Flow = 0)	Speed (m ³ /ks)	Advantages	Disadvantages
Oil-Sealed Rotary Vane	10^{-4}	200	10^9	0.1-10	Wide Range; High Compr.	Not tritium Compatible
Dry Rotary; Dry Piston	10^3	10^3	100	0.1-10	Wide Range; High Compr.	Not Hermetic; Materials Prob.
Metal Diaphragm	$3(10)^3$	10^5	30	1-5	Hermetic; Reliable	Bulky; Expensive
Metal-Bellows	$2(10)^3$	300	15	1-3	Hermetic; Reliable; Compact	Limited Vac. Capability
Dry-Contact Scroll	10^4	100	5	0.05	Compact	Limited Speed & Press. Range
Controlled-Clearance Scroll	10^{-5}	10	10^4	5-100	High Compr.; High Speed; Reliable	Unusable for Transfer above 10 kPa
Bellows-sealed Wobble	10^{-3}	10	1-10	8-140	Hermetic; High Speed	Bulky; Expensive; Limited Comp.
Canned-Motor 2-Stage Roots	10^{-4}	10	700	50	High Speed;	Oil Contam- ination
Air-bearing Turbopump	10^{-7}	100	10^{12}	500	High Speed; High Vacuum	No Transfer Capability
Mag-bearing Turbopump	10^{-7}	10^{-5}	10^5	500	High Speed; High Vacuum	No Transfer Capability

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Fig. 22. Oil-less diaphragm vacuum pump (Courtesy of KNF Neuberger, Inc., Princeton, NJ).

compatible with tritium gas. Diaphragm pumps (Fig. 22) avoid the lubrication problem, and they may be completely leak-tight but their elastomeric or plastic diaphragms would be rapidly embrittled, if used to pump tritium gas.

One can take the diaphragm idea a step further and make it out of metal to provide tritium compatibility. In fact diaphragm compressors have been used at many tritium facilities for many years (Ref. R-5) The best pumps for tritium incorporate three stainless steel diaphragms with the upper diaphragm welded to the pump head (Ref. M-30). The stacked diaphragms are flexed with hydraulic fluid driven by a reciprocating piston. A leak test port between the diaphragms (Fig. 23) yields continuous information on the integrity of the gas

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Fig. 23. Triple metal-diaphragm vacuum pump.

diaphragm and its seal weld. Although this type of compressor has been very successful, its use in the subatmospheric pressure range requires complex design which greatly increases its size and cost, as well as making it much more difficult to maintain than a metal-bellows pump of the same performance characteristics.

Metal-bellows pumps are available from commercial sources (Ref. M-14) and have also been used for many years at some tritium installations (Ref. B-3). These devices are available in all-metal construction with completely hermetic motion transmission (Fig. 24). Compressions are fairly low, but series-staging of these relatively small pumps allows vacuums as low as 10 torr to be produced. Below that pressure gases do not carry enough momentum to operate the reed-type stainless

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Fig. 24. Metal-bellows vacuum pump principle
(Courtesy of Metal Bellows Corp., Sharon, MA).

steel check valves. The manufacturer offers these pumps in many sizes and versions, including double-bellows (secondarily contained) models (Ref. R-4).

The orbiting scroll pump (Ref. 1) is a recent development that makes use of a novel pump geometry. The pumping action of this device will be described subsequently under booster pumps (Sec. E.2). In the version useful in the viscous pressure regime, gas is compressed between two interleaved plates with spiral channels. The plates rub during operation, so they are fabricated from graphite to reduce internal friction. This graphite scroll pump introduces shaft rotation into the pump chamber by a canned magnetic drive mechanism and converts this into the required orbital motion internally by an

Oldham's coupling. This coupling also has friction, which is minimized by fabricating the mechanism out of polyimide. Although not ideal for tritium (both graphite and polyimide retain water and chemically react with tritium to produce tritiated waters and hydrocarbons - see Refs. 4 and R-2), this pump is nevertheless hermetically sealed and truly oil-free. The manufacturer (Ref. M-29) is actively pursuing several designs based on the scroll principle.

One extremely simple device that may be considered for some vacuum roughing applications is the venturi pump (Fig. 25). This "pump" produces a partial vacuum at the expansion throat just behind a venturi nozzle, requiring only a fixed-flow geometry and a source of pressurized gas. These very small units work with all pressurized fluids including hydrogen. They are particularly useful as flow mixers. They can also be useful for rough evacuation where both the driving and

Fig. 25. Venturi type evacuation pump.

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aspirated gases exhaust to a tritium waste treatment system (Part IV of these notes). Inexpensive pumps of this type, some of which can produce vacuum as low as 5 torr, are available from several sources (Refs. M-26, M-31, M-32).

E.2. Booster Pumps

The pressure regime between a few millitorr and a few torr has always been the most difficult for tritium pumping. All high vacuum pumps suitable for tritium (see Sec. E.3) require forepressures below a few millitorr to operate. Since none of the tritium-compatible roughing pumps discussed in the previous section will produce vacuums this low, some type of tritium-compatible booster pump is required to span the transition pressure range (100 millitorr--10 torr). The principal contenders for this job are the canned-motor Roots blower, the "wobble-plate" pump and a new French-built moving spiral pump.

Roots blowers show considerable promise for tritium in the transition pressure regime, as several canned-motor pumps are now available. Even though the canned motor provides true hermetic isolation from the atmosphere, tritium compatibility remains a problem because of incomplete isolation of the gear and bearing lubricant from the working vacuum chamber. There is one report in the literature (Ref. 2) of a Roots pump with Ferrofluid-TM (Ref. M-7) shaft seals. Only one of these small machines was ever built, but the technology could probably be revived and improved for fusion vacuum systems. Another solution to this problem (Ref. 3) is to install essentially

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vacuum-tight seals on the rotor shafts (Fig. 26), and to evacuate the gear case with a conventional vacuum pump. Any tritium leaking past the rotor shaft seals is then exhausted to a waste treatment system. The differential pressure across the shaft packing inhibits aspiration of oil into the pump chamber, where it would contaminate the tritium vacuum medium.

The moving-spiral pump is discussed in the author's previously published paper (Ref. R-9), reprinted here as an appendix. The "wobble-plate" pump is also discussed in this and another reprinted reference R-4. The descriptive information given in these references will not be repeated here.

Fig. 26. A canned-motor Roots pump with evacuated gear case (from H. Bode, Ref. 3).

E.3. High Vacuum Pumps

If diffusion pumps are eliminated from consideration, we are left with only some type of capture pump (discussed in Sec. E.4) or turbomolecular pump to produce our ultimate vacuum in the tritium-handling system. There have been exciting developments in turbomolecular pump technology in recent years. Early turbopumps had neither adequate compressions nor speeds for the light gases like helium and hydrogen. Moreover, tritium compatibility was a problem because of the organic lubrication required for the high-speed bearings exposed to the vacuum medium. Failure of the turbopump power supply could also result in contamination of the high vacuum chamber from diffusion of oil vapor. Turbopumps that overcome these difficulties are now available.

One manufacturer (Ref. M-13) markets a turbopump design in which the high-speed rotor is suspended in vacuum by a magnetic field, thus completely eliminating mechanical bearings with their lubrication requirements. This pump (Fig. 27) has a pumping speed greater than 500 L/s for both hydrogen and helium, and it tolerates forepressures as high as 0.1 torr. As supplied by the manufacturer, this pump contains a few elastomeric static seals, but these are easily replaced with metal wire-rings to achieve complete tritium compatibility.

Another turbopump manufacturer (Ref. M-3) has introduced a small pump (60 L/s) of hybrid design--conventional lower bearings and permanent-magnet upper bearings (Fig. 28). Conventionally lubricated turbopumps have been used successfully

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Fig. 27. Magnetic-bearing turbomolecular pump
(Courtesy of Leybold-Heraeus Vacuum Products, Export, PA).

Fig. 28. Turbopump with permanent-magnet upper bearings
(Courtesy of Balzers, Hudson, NH).

in tritium-containing vacuum systems. The exposure of the lubricant is moderate in the operating pressure range of these pumps. As the more reliable solid-state frequency converters come into use, there is less risk of turbopump failure followed by oil backstreaming into the high vacuum system. Even so it is better to back turbopumps with the oil-free roughing and booster pumps discussed in the previous sections.

E.4. Capture Pumps

A number of devices working on different principles are classified as reservoir or "capture" pumps. This designation is derived from the fact that these devices all reduce the pressure in a vacuum system by collecting or trapping gas molecules on their interior surfaces. The actual mechanism may be electrostatic trapping (ion pumps), chemical reactions (titanium sublimation, getter, and metal-hydride pumps), physical adsorption (sorption pumps), or condensation (cryopumps). The basic vacuum characteristics of all these pumps cause them to be more effective for some gases than for others. Their capture mechanisms may also be reversible for some gases and not for others. The effectiveness and reversibility of such pumps for the hydrogen isotopes is obviously a major factor in selecting a pump for tritium or DT gas. Helium effectiveness must also be considered for fusion vacuum pumps, because helium-3 (from tritium decay) and helium-4 (from DT fusion) are always present in the plasma exhaust gas.

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Sorption pumps (Refs. M-3, M-12, M-13, M-20, M-24, M-25, M-26, M-35) have been used longer than any of the other types of capture pumps. Both charcoal and the various Zeolites, or molecular sieves (Ref. M-38) may be used at liquid nitrogen temperature (77 K) to trap all gases except helium and neon (Ref. 5). These adsorbents have fairly low capacities for the hydrogen isotopes at this temperature, so pumping DT gas is limited to roughing applications and temporary storage of gas from components in need of repair. Differential sorption also offers some possibilities for separation of DT gas from helium (Refs. 6, 7), which sorbs scarcely at all at 77 K, and from condensable impurities, which stay bound at temperatures where DT gas is desorbed--about 100 K. The typical commercial sorption pump (Fig. 29) must be modified by replacing its simple pressure relief device (often a rubber stopper) with one that is leak-tight and tritium compatible. The relief device must also be ducted to a closed container or tritium waste treatment system. A metal rupture disk ducted to a surge tank is the best arrangement for this application. Activated charcoal is a better choice for tritium than is the molecular sieve normally furnished with these pumps. If separation of gases is to be a function of the sorption pump, a flow-through model with long path length can be easily coiled up from a roll of standard annealed copper tubing (Fig. 30).

The ion or getter-ion pump was developed in the 1950's following the discovery that operating hot-filament ion gauges produced significant pumping. The principle is illustrated in

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Fig. 29. Standard sorption pump (Courtesy of Perkin-Elmer Vacuum Products, Eden Prairie, MN).

Fig. 30. A simple flow-through sorption trap (Courtesy of Los Alamos National Laboratory, Los Alamos, NM).

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Fig. 31. Ion pumps are effective for all but the inert gases, and they have enhanced speeds for the hydrogen isotopes in particular. The inert gases, helium and argon, can also be pumped in limited quantities with specially designed getter-ion pumps. The gettering action of ion pumps is greatly enhanced by evaporating titanium, in which case they are called titanium sublimation pumps (TSP's). With getter-ion or titanium sublimation pumps, the gases are pumped irreversibly, and when the getter material is exhausted, the pump must be replaced with a fresh one. These pumps are useful for maintaining very high vacuums on cryogenic insulating jackets where the gas load is low. Obviously they cannot be used where DT gas is a significant part of the gas load, because the tritium is not recoverable.

↔(a) Conventional

↑
(b) Differential (Argon-stable)

Fig. 31. Principles of getter-ion pumping (Courtesy of Perkin-Elmer Vacuum Products, Eden Prairie, MN).

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At tritium facilities throughout the United States uranium hydride beds have long been used for storage of tritium and evacuation of tritium supply containers. The ready availability of depleted U-238 at the national nuclear laboratories is one reason for its popularity. Another reason is evident from the phase diagram of the uranium/hydrogen/uranium-hydride system (Fig. 32). Uranium will getter DT gas to pressures as low as 0.001 millitorr at room temperature, and yet the dissociation pressure is a convenient 760 torr at just over 400 deg C. Moreover, oxygen, nitrogen and carbon are gettered irreversibly by uranium, so the hydrogen isotopic mixture can be purified each time it is cycled through the uranium hydride phase. Uranium hydride beds are not commercial items, but each

Fig. 32.
Dissociation pressures
of the 3 hydrogen
isotopes over their
hydrides of uranium.

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tritium facility has its favorite design. The Los Alamos model (Fig. 33) features multicolumn flow-through design. The uranium container is vacuum-cast copper to maximize heat transfer and minimize DT permeation. All heaters and thermocouples are mounted in nonconnecting bores in the copper block. With evacuated superinsulation surrounding the copper block, it can be maintained at an operating temperature of 450 deg C with only a 200-watt heater. The massive copper block can absorb all the exothermic heat from hydriding the entire charge of uranium with a temperature increase of <100 deg C. If stainless steel is used for the bed container, extreme care must be exercised to prevent inadvertant overheating, as an iron-uranium eutectic occurs at 750 deg C.

Fig. 33.
An all-copper flow-through
uranium getter bed design
(Courtesy of Los Alamos
National Laboratory).

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One severe disadvantage of uranium is its physical form after hydriding. It breaks down into a micron-size powder which is very difficult to confine to its original container, particularly during the viscous flow conditions encountered during dissociation. Besides being highly pyrophoric, this fine powder plays havoc with valves, mechanical pumps, and the tritium handling system in general. Although none of them have the convenient working temperature range of uranium, newer alloys have been developed and marketed (Refs. M-21, M-33) that maintain their physical form during repeated hydriding and dehydriding. These should be seriously considered for DT gas storage, even if some other mechanism must be used for obtaining high vacuum.

Non-evaporable getter pumps (Ref. M-21) that use zirconium-aluminum alloys are used extensively in the fusion program. The pumps, which are available as nude cartridges, panels, or enclosed appendage pumps (Fig. 34), have high speeds and useful reversible capacities for the hydrogen isotopes. They also getter irreversibly reasonable quantities of impurity gases (nitrogen, oxygen, etc.), so the DT gas recovered from these getters is purified in the process. Like uranium, these getters will not pump any of the noble gases. Unlike uranium, the Zr-Al getter maintains its physical shape during hydriding and dissociation--that is, it does not powder, provided that its recommended capacity is not exceeded. These pumps can be used to getter vacuum jackets surrounding high-temperature, hydrogen-permeable components, because they can

Fig. 34. Non-evaporable getter pump cartridge (Courtesy of SAES Getters S.p.A., Milano, Italy).

be sized to keep up with high gas permeation rates, the gettered DT gas is recoverable, and the getter cartridge is reusable.

Cryopumps have become very popular since the 1960's. Their much-advertised advantages include low cost, high pumping speed, hydrocarbon-free vacuum, and easy scaling to large sizes. Commercial cryopumps, operating from closed-cycle helium refrigerators, are available from many sources (Refs. M-3, M-13, M-20, M-26, M-34, M-35, M-36, M-37). Because general cryopumping is the subject of another AVS short course, further discussion here will be limited to tritium cryopumping applications only.

The refrigerators furnished with laboratory-type cryopumps do not actually liquefy helium, so their cryopanel operating temperatures are limited to about 10 K and above. At this temperature helium does not condense at all, and hydrogen has a vapor pressure >1 torr, so some mechanism other than condensation is required to produce ultra-high vacuum when these gases are present. The commercial pumps above solve this problem by bonding a small amount of adsorbent (usually charcoal) to the underside of the cryopanel. Since these pumps are intended to produce and maintain vacuums in chambers originally filled with air (containing only 5 ppm helium and 0.5 ppm hydrogen) they do not normally contain enough sorbent to handle plasma exhaust gases, comprising hydrogen and helium isotopes almost exclusively.

Despite the apparent shortcomings of commonly available cryopumps for the light gases, large-scale cryopumps operating at liquid-helium temperature (4.2 K) are currently the best prospects for plasma-chamber evacuation at fusion power facilities. In such machines the hydrogen isotopes (DT gas) are evacuated by condensation on smooth metal panels refrigerated to 4.2 K. Even at this temperature helium must be physically adsorbed to reduce its pressure to the high vacuum regime. Three helium cryosorbing materials have been studied and tested to some extent (Ref. R-8); they are 5A Molecular Sieve, activated charcoal, and argon frost deposited on the cryopanel concurrently with the sorbed helium. The first two materials will not adsorb helium in the presence of significant DT, so a

"compound" pump had to be designed. A compound cryopump is one in which the helium-cryosorbing surface is shielded from the incident DT by the DT-condensation panel itself, usually a set of chevrons or other optically opaque baffle (Fig.35).

If cryopumps have so many advantages, and if they can be designed and built to pump even the plasma exhaust gases, it is reasonable to ask why any other type of vacuum pump is needed around a fusion plant. The answer lies in the fact that a cryopump is a reservoir-type device with a quite finite capacity when compared to the gas loads coming out of a power-producing tokamak reactor. When this capacity is reached, usually after a few hours, the pump must be closed off, the cryopanel allowed to warm slightly, and the vaporized gases

Fig. 35.
The Los Alamos
compound cryopump.

pumped away. Only then can the panels be refrigerated again to restore the cryopump for another useful pumping cycle. Of course the plasma cryopump could be regenerated into another cryopump, and so forth, ad infinitum. The chemical fuel purification and isotopic separation processes require steady gas flows to function properly however, and the batch surge flows resulting from successive cryopump regenerations does not provide this. Therefore some combination of mechanical pumps, chosen from those discussed in the previous sections, must be used during this regeneration process to remove the vaporized and desorbed gases.

Compound cryopumps have an additional advantage when used in a fusion plasma evacuation system. Since the helium "ash" must be separated from the recycling DT fuel during the purification process, and since He and DT are collected on separate cryopanel, it is possible to effect this separation in the cryopump during a controlled regeneration. "Controlled" is the operative term here, and it requires extremely delicate and separate temperature control on each cryopanel, and maintenance of vacuums below 0.1 millitorr all during the regeneration process. Otherwise the panels warm too quickly, releasing the gas faster than it can be pumped away. This results in an instability known as cryogenic runaway, and all the gases on both panels vaporize and mix almost instantaneously. Despite these difficulties, this capability of cryopumps has been demonstrated (Ref. R-8), but this design feature places further restrictions on the choice of the pumps.

Typical Gage Readings
(torr) at Zero
Throughput (Blank-off).

Fig. 36. A tritium-compatible cryopump-regeneration system
(from Ref. R-8).

for regeneration, namely in terms of the high vacuum pumping speeds required. A cryopump regeneration system (Fig. 36), comprising a series of tritium-compatible mechanical pumps, is described in appended reprint R-9. This discussion on cryopumping is concluded by referring to the extensive literature in references B-9, B-10, B-12, and B-13, and in reprints R-4, R-8, and R-9, appended to these notes.

E.5. Service Vacuum Systems

After the vacuum and processing systems for any facility have been designed, there always remain a number of uncritical evacuation functions that need to be provided regularly or occasionally. A service or "house" vacuum system is normally installed for such purposes. The most common installation is a large oil-sealed rotary-vane or rotary-piston vacuum pump coupled to a large manifold, routed throughout the facility to a number of valved "service" evacuation ports. Uses for such a system at a fusion facility include evacuation of glove box antechambers (pass boxes), initial and postmaintenance rough pumping of vacuum jackets on large in-place components, etc.

The safest assumption to make regarding a house vacuum system serving a tritium facility is that the system WILL become tritium contaminated. All design should be contingent upon this assumption, even though operating procedures and administrative rules forbid use of the system for "hot" components. This philosophy anticipates the presence tritiated water, oil, and contaminated components whenever the system is subsequently broken into, even for routine maintenance.

Because oil and tritium are basically incompatible, an oil-free pump should be chosen for the house vacuum system. Ten years ago this was not possible, but now the larger models of available dry moving-spiral pumps (Ref. M-17) can provide adequate vacuum and good speeds for house systems. A large Roots blower, backed by an oil-free piston or diaphragm fore-pump, is another choice open to designers today. There is no

logic favoring the expenditure of thousands of dollars necessary to install all oil-free pumps in the main tritium processing systems, and then exhaust all the tritium-contaminated waste streams through the only oil-filled vacuum pump in the facility. Nevertheless, this is the usual practice, largely because preexisting house systems are considered "good enough," and the assumption about tritium contamination is denied. Besides creating a radiological exposure problem for operating personnel, the exposure of oil to tritium puts an additional burden of tritiated hydrocarbons (HC's) on the cleanup system (Part IV). The pump's exhaust must be filtered for oil mist, and the vacuum inlet line baffled against back-diffusing oil vapor. No filters for either of these functions are completely effective. The paragraphs that follow do assume that oil-sealed pumps are used for house vacuum systems, in recognition of the fact that vacuum designers also live in the real world.

Although any oil-filled pump will lead to contamination problems in a tritium facility, some types are worse than others and should be avoided. Direct-drive pumps are preferable to belt-driven models of equivalent performance. Belts break frequently even in normal service, but they also "throw" the oil that inevitable leaks or seeps past the pump shaft seals. Since this oil is tritium contaminated (our fundamental assumption), the high speed belt immediately spreads this contamination over a wide area. Many manufacturers make direct-drive pumps with special high-integrity or separated

duplex seals (Refs. M-2, M-13, M-26). These are known as "hermetic", "helium-3", or "exotic gas" models, and house vacuum systems at tritium facilities should make use of these models, even though they are considerably more expensive. In actual fact, if conventional pumps are replaced with these types before the system becomes contaminated, the extra cost of the hermetic pump will probably be earned back in reduced radiological exposures the first year, thanks to much more efficient containment of tritiated oils.

Before placing any oil-filled pump into service it should be fitted with valves on the oil drain and fill ports. This enables service personnel to change the oil with minimum contact. Dirty oil should always be aspirated into a preevacuated waste container filled with sorbent (vermiculite or diatomaceous earth) to immobilize the contaminated oil before disposal by burial. The charge of fresh pump oil can be siphoned into the pump by a similar "hands-off" scheme. Double-shutoff quick-disconnects on the pump, oil drain container and refill container greatly facilitate this operation, which is shown schematically in Fig. 37 and described in more detail in the appended reprint R-6.

The rotary pump should be installed so that routine maintenance (oil changes, etc.) can be performed without disconnecting inlet and exhaust piping. It should also be set in a liquid-tight tray or tub of sufficient volume to hold the pump's entire oil charge in case of catastrophic seal failure. The best state-of-the-art coalescing oil-mist filter should be

Fig. 37.
No-contact method for
changing vacuum pump oil.

installed on the discharge, and a passive anti-diffusion vapor trap (Molecular Sieve or wire mesh) should be installed in the inlet line leading to the vacuum manifold. These devices will retard the migration of oil away from the pump, though they will not stop it entirely. The filters and traps should be of the throw-away type, and should be installed with the 3-valve system shown in Fig. 38. Since the filters will be soaked with tritium-contaminated oil when they are no longer serviceable (our basic assumption again), the valves protect personnel and the environment during the changeout. The entire filter with its valved off housing, not just the cartridge insert, should be sent to waste burial.

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Fig. 38. Maintenance valve arrangement for replacing contaminated tritium system components.

Finally, since the exhaust of the vacuum pump is assumed to contain tritium in some form (HT, HTO, or tritiated HC's), it must be contained and ducted to the facility waste treatment (tritium-removal) system. Many such systems have a partially evacuated holding tank as their first component, and most oil-sealed pumps will not operate properly when discharging to subatmospheric pressure. Therefore a back-pressure regulator, set at 1 or 2 psig, should be installed between the pump's discharge and the waste treatment inlet manifold in such systems. The pressure drop across the regulator allows both the vacuum pump and the waste treatment system to operate within their rated pressure ranges.

F. Secondary Containment of Vacuum Systems

Over the years a modus operandi has evolved for controlling escape of tritium into work areas and the environment. A principal feature of this method is that tritium gas is always shipped and stored at subatmospheric pressures in double walled vessels. The expectation is that even if there are small leaks in the primary vessel, no radioactive tritium will leak out, because the differential pressure will cause in-leakage of atmospheric air instead. By the same reasoning there should be no requirement to secondarily contain tritium-pumping vacuum systems, because vacuum equipment works in the subatmospheric regime by definition. There are, however, some difficulties with this strategy.

In the first place, in a fusion tritium-handling facility like the Los Alamos Tritium Systems Test Assembly (TSTA) there is no sharp, well defined interface between vacuum processes and pressurized operations. Second, the inleakage of atmospheric air into the tritium process gas creates an operational problem and constitutes an even more serious radiological hazard than does external leakage of tritium. This is because the oxygen leaking into concentrated tritium is converted to the oxide very rapidly. The oxide conversion rate is so high compared to any reasonable leak rate that within a few hours secular equilibrium is established between the oxygen leaking into the container and that converting to tritium oxide. The same is true for conversion of ordinary water vapor to the tritiated species by hydrogen isotopic exchange. The result

of both processes is a continuous production of tritiated water and a simultaneous reduction of tritium gas at a molar rate at least double that of the oxygen inleakage rate, because of the chemical reactions:

The reason these reactions proceed so rapidly in the situation described is that the concentrated tritium gas creates an intense ionization field due to its radioactivity, and this catalyzes any exothermic chemical reaction possible with available constituents. From the above you can see that tritium converted to oxide is removed from the useful inventory just as surely from air inleakage as from tritium outleakage. Tritiated water is also thousands of times more hazardous radiologically than is an equivalent quantity of elemental gas.

By contrast if any tritium leaking out of the system is simply allowed to disperse to its equilibrium concentration in the atmosphere, it is unlikely to convert to oxide spontaneously for years, and it will certainly be very dilute by then. (See the author's paper, appended reprint R-10, for extended discussion of this point).

There are two main points to be made from all this discussion: First, operating below atmospheric pressure is neither a substitute for a leak-free system, a guarantee against loss of tritium, nor a panacea for radiological exposure; second, if secondary containment is employed, the containment medium should not be air, and especially not wet air.

Some of the vacuum pumps discussed in the previous sections can be purchased with integral secondary containment features, and the most failure-susceptible component of bellows valves can be protected (Fig. 18). Some of the absolute and differential pressure gauges described in Sec. D also have intrinsic secondary containment construction in the geometry of their reference chambers. Because none of these components have infinite lifetimes, nor are they totally maintenance free, they must be installed with reconnectable fittings like those described in Sec. B. As good as these are, they all have finite and cumulative leak rates, and a typical small system with pumps, valves, gauges, etc., can easily use more than 100 fittings. The simplest and least expensive way to provide secondary containment for everything is to install the entire system in a glovebox. As one example of the cost effectiveness of this approach, a 1-cubic-meter glovebox may be purchased (Ref. M-41) for the extra money that would have to be spent for a double containment version of one small metal-bellows pump (Ref. M-14). The glove box also provides containment for all the auxiliary components and fittings. (Fig. 39).

Stainless steel has been the choice of the nuclear fuel-processing industry (fission) for glove box construction, because of its easy-to-clean surface and resistance to attack by chemical agents. For processing tritium, which does not require wet chemicals, aluminum is just as satisfactory for gloveboxes, and it offers an almost 50% cost advantage. Aluminum is also lighter, and it is more amenable to field-built

Fig. 39. Simple bellows transfer pump system enclosed in a controlled-atmosphere glovebox.

penetrations and modifications. Each glovebox should be fitted with a double-door airlock (antechamber) large enough to pass through the largest component installed in the box. The gloves in a box that houses a remotely operated system are for performing "remove-and-replace" maintenance rather than for actual manual operation of components, and this philosophy should guide the placement of intrabox components. An extensive and growing literature on glovebox and glovebox operations already exists (Refs. B-21, B-22), so further discussion here is limited to the specific problem of containing tritium.

The choice of glovebox atmosphere has been considered in conjunction with the cleanup system, which is the subject of Part IV. Argon is a good choice because it is inert, is easily separated from the hydrogen isotopes by physical means, and it

behaves pretty much like air thermally and mechanically. Helium is sometimes used because of its superior heat-transfer characteristics, and because leak detection of a helium-filled glovebox is a straightforward operation with a helium-sensitive leak detector. Any glove box containing tritium-gas process piping will require frequent purging to maintain the tritium below an acceptable concentration, and it is not economical to release this much argon or helium to the atmosphere. Glove box systems filled with these gases must therefore be operated with a recirculating cleanup system.

At TSTA dry nitrogen was chosen as the glovebox medium, and the waste treatment system is a once-through process with the tritium-free nitrogen exhausted to the atmosphere. Nitrogen is essentially inert to tritium at ambient conditions, and no water is synthesized from tritium leaking into the glovebox. Other advantages of nitrogen are that liquid-nitrogen dewars required for cryogenic processing and nitrogen-operated pneumatic valves can be vented directly into the glove box without changing the composition. In fact these additional sources of gas supply some degree of automatic purging. Nitrogen is abundant and only slightly more expensive than compressed air. In a facility requiring lots of liquid nitrogen, and this certainly includes all fusion reactors currently under study, adequate supplies of dry nitrogen for glovebox filling and purging are available from dewar and transfer line boiloff, and this gas is essentially free.

Where tritium lines must be run from one glovebox to another, they should be run within a secondary jacket. This jacket needs to be gas-tight and should be filled with the glove box medium. The ISO-KF flange system provides a convenient means of assembling this secondary conduit, and this application is illustrated in Fig. 40.

Fig. 40. Glovebox penetration for tritium piping, complete with secondary containment jacket.

IV. TRITIUM WASTE TREATMENT SYSTEMS

A. Sources of Tritium and Release Limits

Because of its high cost and radiological impact on the environment, tritium must be meticulously confined in all parts of a fusion plant. This means using high-integrity primary piping plus additional levels of containment, anticipating possible escape routes, and physical or chemical recovery of any significant amounts converted to nonelemental form during processing. There are multiple sources of low-level tritium which must therefore be intercepted to keep the environmental impact as low as possible.

Tritium dissolves and diffuses into all metals, and it reacts with most elements, so low levels of tritium or tritiated compounds will be encountered in the exhausts of all analytical instruments, vacuum pumps, and vacuum systems serving a tritium facility. Because service or "house vacuum" pumps are normally conventional oil-sealed rotary pumps (see Part III, Sec. E.5), tritium from these sources will be exhausted mainly in the form of tritiated water vapor and tritiated hydrocarbons (HC's).

A second major source of tritiated effluent is the gas purged from the facility gloveboxes and other secondary-containment volumes. The tritium in this gas is the result of leakage, permeation, or accidental releases from the primary system, so it will most likely be deuterium-tritium (DT) in

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elemental form, diluted with whatever gas constitutes the glovebox atmosphere.

At any major DT-fuel-processing facility, like the Los Alamos Tritium Systems Test Assembly (TSTA), a number of gaseous waste streams are generated in the course of purifying the plasma-refueling DT. Although all the recoverable DT is removed from these waste streams they must still be treated to reduce tritium concentrations to the lowest possible levels before discharging to the atmosphere. These waste gases include the He-4 produced from the DT fusion itself, He-3 from radioactive decay of tritium, normal atmospheric or glovebox gases resulting from inleakage, and common or exotic gases resulting from interactions of the fusion plasma with its container. Some processes may create other gas streams as well; an example in the plasma evacuation system is argon-frost cryosorption (Batzer, et al--Ref. B-13), which yields significant quantities of argon gas during cryopump regenerations.

A system for routine tritium removal must be available to receive and treat all the above waste streams before they can safely be discharged to the atmosphere. In a power-producing fusion plant, economic considerations coupled with the discharge rate of the noble gases (Ar, He-3, and He-4) may dictate that these also be separated, purified and recovered. In any case it is feasible to remove tritium from all these waste streams by either a continuous or frequently cycled batch system. Fig. 41 is a schematic of the sources and discharges of a waste treatment system at a typical fusion facility.

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Fig. 41. Tritiated waste streams at a fusion power facility.

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One source of tritium effluent that cannot be so conveniently ducted to the routine waste treatment system is that resulting from an accidental release into the building itself. A catastrophic failure or natural disaster that breaches both the primary and secondary piping is one example; another is a release of tritium from a shipping container during load-in or load-out operations, when there may be a temporary discontinuity in the secondary containment. To accommodate these basically unplanned releases into the facility, large emergency cleanup systems, capable of processing and cleaning up the entire facility atmosphere, are being considered for fusion facilities. These systems are also discussed in Sec. C.

There are no hard-and-fast release limits for tritium, but all modern tritium facilities are being designed to meet radiogenic exposure criteria imposed by the Nuclear Regulatory Commission (NRC) on light-water-cooled fission reactors. There are also Dept. of Energy regulations (DOE Order 5480.1A, Chap. XI) that set limits for exposures to facility workers and to the general public. Both DOE and the NRC regulations, which apply to all types of radiation, limit exposures to radiation workers to 5000 mrem/yr. DOE limits for the general public are 500 mrem/yr for an individual permanently at the point of maximum probable exposure (site boundary,) and 170 mrem/yr per individual for any significant population. NRC limits for reactor-site radiation are lower--25 mrem/yr per individual. NRC regulations also set total doses (man-rem/yr) for populations within a certain radius of a reactor site.

None of these exposure guidelines are easily translated into stack release rates for tritium. The exposure to populations is a function not only of the release rate, but of many other factors, most of which are continually changing. These factors include stack height, local geography, emission concentration, fraction of tritium oxidized at release time, prevailing weather at the time of the release, and diurnal shifts in population density and distribution.

Another principle adopted for DOE-funded construction projects is that releases of radioactive materials to the biosphere and resulting exposures to the public shall be "as low as reasonably achievable" or ALARA. Although dose rates to the general public are difficult to calculate for any except purely hypothetical conditions, tritium-control technology has now advanced to where the ALARA principle yields exposures to members of the public lower than those permitted by either DOE or NRC stipulations. The main elements of this technology are secondary and tertiary containment and the chemistry-based waste-stream treatment systems discussed in the sections that follow.

B. Principles of Tritium Removal

The design of tritium waste-treatment or tritium-removal systems is usually the responsibility of a chemical or a mechanical engineer. The vacuum engineer should at least understand the basic processes however, because all sources of tritium-contaminated waste, including vacuum system exhausts, do impact the design of the cleanup system. In its simplest terms tritium cleanup consists of oxidation of all the hydrogen isotopes, including tritium, to their respective waters, followed by collection of the tritiated water on a high-efficiency dessicant before discharging the dried and tritium-free waste gases up the stack.

Because the hydrogen isotopes are extremely dilute in the waste streams, a catalyst bed is provided to increase the interactive dwell-time between hydrogen and oxygen. All operating cleanup systems to date have used either (1) a family of proprietary catalysts (Ref. M-42) containing platinum (Pt) and/or palladium (Pd) on an alumina or molecular-sieve substrate; or (2) a mixture of copper oxide and manganese dioxide marketed under the name of Hopcalite (TM of Mine Safety Appliances Co.--Ref. M-43). The Hopcalite catalyst has an advantage for treatment of continuously recirculated inert gas streams because it provides the oxygen required for the conversion to water. If a surplus of oxygen already exists or can be easily injected, the Pt-Pd catalysts are usually more efficient (have a higher decontamination factor) at any given operating temperature.

Dessicants (water adsorbents) that have been used include silica gel, activated alumina, and the molecular sieves, with the last-named overwhelmingly preferred by waste-treatment system designers. Molecular sieves have very high capacities for water and can dry air to less than 1 ppm at ambient temperatures. They can also be tailored to adsorb substances other than water, hydrocarbons for example. They are readily dried (regenerated) in place by purging them with hot gases, and thus they can be used over and over almost indefinitely.

It would appear that the vacuum engineer and tritium processing personnel need only tell the waste-treatment system designer what flow rates and tritium concentrations to expect, and straightforward chemical engineering calculations will lead to the design of an adequate cleanup system. Of course there are some other factors to consider, and these require close coordination among all the designers involved. Some tritium in the waste streams will not be in the elemental form, and in particular, hydrocarbons like methane, ethane, and acetylene can be expected. These HC's will be more abundant if oil-sealed pumps are used to evacuate tritium-contaminated components and move the waste streams. Both catalysts discussed above must be operated at elevated temperatures in order to "crack" and oxidize the HC's efficiently. Both catalysts are poisoned by halogens and by carbon monoxide, and a number of other substances, including mercury and sulfur compounds, have deleterious effects on the preferred Pt-Pd catalysts. This is

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additional justification for avoiding mercury or halogenated pumping fluids in any vacuum system exposed to tritium

Adequate data exist both for catalysts and dessicants to design cleanup systems for any desired performance, given the incident gas loads, concentrations, and flow rates. Several companies (Refs. M-41 through M-45) offer everything from individual components to complete system design and fabrication. Folkers and Cena (Ref. 9) give a brief but comprehensive survey of the trade-offs involved in designing such systems. This reference also includes an extensive bibliography on cleanup system designs and performance tests.

For inert gas waste streams (some glovebox atmospheres, for example) another type of cleanup system based on hydrogen-selective gettering is possible. This scheme has the advantage that it does not create large quantities of dilute, but still radiogenically hazardous, tritiated water. Furthermore, since all the materials considered getter hydrogen reversibly, the DT extracted from the waste stream can be recovered by heating and subsequently restored to the fuel loop. Getter materials considered to date have limited capacity for oxygen and nitrogen, so they are probably not practical for gloveboxes, where air inleakage and permeation are facts of life. Titanium can getter these atmospheric gases (Ref. 13), so a combination of different getter beds may be a solution. Such systems have been proposed (Refs. 10, 12), but so far as the author knows, none are actually in use yet for tritium facilities.

C. Typical Cleanup System Designs

In this section cleanup systems of several types likely to be used at a fusion plant will be briefly described. Unless totally new catalysts, dessicants, or hydrogen-selective getters are developed, refinements and scale-ups of these basic designs will be principal features of any fusion facility planned during the next decade or two.

C.1. Continuous Cleanup Systems

A Gas Purification System (GPS) purifies the glovebox atmospheres at the Tritium Research Laboratory (TRL) of the Sandia Laboratories, Livermore, CA (Ref. 11). This system, which was built by one of the systems manufacturers (M-42) to Sandia Laboratories' performance specifications, is shown schematically in Fig. 42. It is typical of the continuous-flow cleanup systems designed for tritium-glovebox atmospheres, though it can also be operated in a batch mode. The glovebox atmosphere at this facility is normally dry nitrogen, but the chemical cleanup system is designed to work just as well with noble gases or even air. Since this system does use the Pt-Pd catalyst, oxygen must be added if the atmosphere is inert.

One feature of this system not mentioned previously is the water swamping between the two series-arranged dryers. After the first dryer reduces total moisture in the exit gas stream to 1 ppm, the gas is resaturated with ordinary water (steam) and passed through the second dryer, so any tritiated water still present is further diluted by several orders of

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Fig. 42. The Gas Purification System at the Tritium Research Laboratory (Courtesy of Sandia Laboratories, Livermore, CA).

magnitude. This is more important for a single pass cleanup system than for this recirculating one.

A second type of continuous cleanup system is illustrated by the Glovebox Atmosphere Detritiation System (GADS) at Mound Laboratory's Tritium Effluent Control Laboratory (Ref. 15). Instead of catalytic oxidation, this system (Fig. 43) uses selective adsorption to remove tritium and other impurities from a helium glovebox atmosphere. Adsorbent columns at different cryogenic temperatures adsorb normal air components and the hydrogen isotopes. The adsorbent beds can be warmed up independently; the air components are sent through a separate batch-type waste-treatment system before they are exhausted to

Fig. 43 A glovebox atmosphere detritiation system (Courtesy of Mound Laboratory, Miamisburg, OH).

the stack. The hydrogen isotopes are not deliberately oxidized in this recovery scheme, so they may be collected during bakeout of their adsorbent bed and reinjected into the fuel processing loop for nearly complete recovery of the DT.

Yet a third continuous-flow system might be based on the reversible hydrogen-selective metal getters (Ref. 11). Such a system would require circulation of the inert gas (He or Ar) through a high-temperature packed bed of proprietary getter alloy (Fig. 44). Although this getter will also capture small amounts of normal air impurities (nitrogen, oxygen, carbon dioxide, methane, etc.) its gettering of the hydrogen isotopes is completely reversible. It therefore shares the virtues of the Mound Laboratory GADS of 100% recovery of any DT gas removed from the glovebox atmosphere. Although this proposed

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Fig. 44. Glovebox detritiation device based on hydrogen getters (Courtesy of SAES Getters, S.p.A., Milano, Italy).

system can purify only noble-gas atmospheres, one manufacturer (Ref. M-21) now offers a getter that will purportedly getter DT from a nitrogen atmosphere. Organic "getters", which are actually unsaturated hydrocarbons like linolenic acid (Ref. 16), have also been proposed for inert-atmosphere cleanup, but their usefulness has not yet been proven.

C.2. Batch Systems

Batch systems can be designed in sizes to accommodate the exhaust of a single vacuum pump or all the diverse waste discharges from a full-scale fusion facility (Fig. 41). Because most batch systems must work effectively on effluents containing both inert and active gases, they are normally designed

Fig. 45. Small batch tritium-removal system for treating a single vacuum pump exhaust (Ref. R-11).

around the catalyzed oxidation process already described. An extremely simple batch system (Ref. R-11) is illustrated in Fig. 45. The entire system was assembled on a 30-cm by 60-cm board. The catalyst was a Deoxo (TM of Engelhard Industries Div.--Ref. M-42) hydrogen purifier, and the operation was initiated by an ordinary pressure switch and sequenced by a simple mechanical programmer.

A more elegant computer-controlled system, using the same basic process, has evolved as the general purpose tritium waste treatment system (TWT) at the Los Alamos TSTA. This system, shown as a block diagram schematic in Fig. 46, has a continuous processing capacity of 60 scfm and can process all the waste streams from TSTA, which is a full-scale prototype of a fuel-handling system for a power-producing tokamak. The actual system is quite complex due to redundant flow paths through all major components, computer-controlled valving, and

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Fig. 46. The TSTA general purpose tritium waste treatment system (Courtesy of Los Alamos National Laboratory).

extensive analytical and radiation instrumentation. This system has provisions for hydrogen and water swamping, oxygen addition if needed, partial or complete recycle, auxiliary input storage, and in-place regeneration of the dessicant. The tritiated water driven off during regeneration is collected on prepackaged moisture collectors for disposal.

C.3 Emergency Cleanup Systems

Despite the relatively small fuel consumption rate at a fusion power plant (5-10 g/hr), the minimum inventory in the fuel processing loop will total several kg of tritium. This is largely due to the inventories required to maintain steady

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state purification in cryogenic components (distillation columns and cryopumps). Because no one can guarantee against catastrophic equipment failures or natural or man-caused disasters, tritium inventories at fusion facilities must be protected against even these events insofar as that is possible.

The Emergency Tritium Cleanup (ETC) at the Los Alamos TSTA is the first major room-cleanup system to be installed as part of a tritium facility anywhere in the world. A complete description of this system is given in appended reprint R-12. A simplified schematic is shown here as Fig. 47. This is a basic catalytic oxidation plus water collection system, but it has unique design features deriving from its intended use.

Fig. 47. The Emergency Tritium Cleanup (ETC) system for TSTA (Courtesy of Los Alamos National Laboratory, Los Alamos, NM).

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First, since this is a standby system that with good luck will never be required during the life of the facility, the catalyst bed is not heated, as doing so would require continuous power consumption. Since any significant release to the building environment will almost certainly be elemental DT gas, oxidation will be quite efficient with the catalyst operating near ambient temperature. Adiabatic heating from the circulating compressor will raise the temperature enough to prevent moisture from adsorbing on the catalyst and blocking its action. Additional heat can be obtained from catalytic burning of normal hydrogen, available by optionally swamping the catalytic reactor input.

The second major departure from usual practice is the condensation and collection of most (90% by design) of the oxides as liquid water in condensate tanks. The rationale for this is the eventual recovery of a significant fraction of the DT lost during a major accidental release. The condensate is segregated into high- and low-concentration batches as it is collected sequentially, and the high-concentration batch can be subsequently electrolyzed into H-D-T and oxygen. The oxygen is stacked, and over a long period of time the hydrogen isotopes can be mixed gradually into the feed stream of the TSTA cryogenic distillation system. Thus the DT can be recovered and the ordinary hydrogen impurity is harmlessly discharged.

Because this is designed as a one-shot system, there is no provision for regenerating the desiccant in place. The used desiccant is simply drained out of the towers and replaced

with a fresh charge. Liquid water from the low-concentration condensate tank is siphoned into the disposable container and adsorbed onto the discarded dessicant.

Another emergency room cleanup system based on slightly different operations is illustrated in Fig. 48. This is the Emergency Containment System (ECS) designed and installed by Mound Laboratory at their Tritium Effluent Control Laboratory (Ref. 15). Again, this is a recirculating, catalytic oxidation system. The principal difference between the Mound ECS and the Los Alamos ETC is in the handling of the tritiated water. In the Mound ECS the heavy-isotope waters synthesized in the catalytic bed are passed through a single large silica-gel bed, already presaturated with ordinary water. The tritiated waters gradually displace the normal water on the dessicant,

Fig. 48. A room cleanup system using presaturated dessicant (Courtesy of Mound Laboratory, Miamisburg, OH).

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and the cleanup is completed before any breakthrough of tritiated water occurs. Here again, the contaminated dessicant is loaded out, disposed of, and relaced, following any accident for which this system must be used. Of course there is no economical way to recover DT from the saturated dessicant in this system.

One generally observed rule-of-thumb is that room cleanup systems should be capable of processing the atmosphere at approximately 1% of the room volume each minute. Even for major releases this cleanup rate enables the tritium concentration to be reduced within 12 to 24 hours to maximum permissible concentration (MPC) for radiation workers. Although data on this subject are very sparse, indications are that no severe water retention problems on building surfaces will result if cleanup proceeds on this schedule. As these systems (ECS and ETC) are built and tested, there are many large-scale decontamination studies that need to be performed.

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The following firms manufacture many vacuum components and equipment that can be used as-is or slightly modified for tritium service. Most of these companies have exhibits at the national and regional AVS symposia, and much useful illustrated literature may be obtained directly from them. Products of particular interest to participants in this course are listed in parentheses following the company address.

- M-1. Advanced Products Co., 33 Defco Park Road, North Haven CT 06473 (metal O-rings, C-rings, K-seals, etc.).
- M-2. Alcatel Vacuum Products, 60 Sharp St., Hingham, MA 02043 (fittings; flanges; Roots, rotary, & turbomolecular pumps)
- M-3. Balzers, 8 Sagamore Park Road, Hudson, NH 03051 (flanges, fittings, Roots and turbomolecular pumps).
- M-4. Cajon Co., 9760 Shepard Rd., Macedonia, OH 44056 (metal-sealed vacuum fittings).
- M-5. Ceramaseal, Inc., New Lebanon Center, NY 12126 (hermetic electrical feedthrus, metal-ceramic tube transitions).
- M-6. Enviropak, Inc., 110 Maryland St., El Segundo, CA 90245 (metal seals for AN-type transducer connectors).
- M-7. Ferrofluidics Corp., 40 Simon St., Nashua, NH 03061 (magnetic-fluid rotary "hermetic" seals).
- M-8. Gast Manufacturing Corp., P.O.Box 97, Benton Harbor, MI 49022 (oil-less vacuum pumps--several types).
- M-9. Granville-Phillips Co., 5675 Arapahoe Ave., Boulder, CO 80303 (vacuum hardware, all-metal valves).
- M-10. Helicoflex Co., 400 Myrtle Ave., Boonton, NJ 07005 (all-metal, spring-loaded, large flange seals).
- M-11. HPS Corp., 1898 Flatiron Ct., Boulder, CO 80301 (vacuum hardware, all-metal valves).
- M-12. Huntington Mechanical Laboratories, Inc., 1010 L'Avenida, Mountain View, CA 94043 (flanges, feedthrus, sorption and ion pumps)
- M-13. Leybold-Heraeus Vacuum Products, Inc., 5700 Melon Rd., Export, PA 15632 (vacuum hardware, magnetic-bearing and conventional turbopumps, Roots and cryopumps).
- M-14. Metal Bellows Corp., 1075 Providence Hwy, Sharon, MA 02067 (metal-bellows vacuum pumps, metal-bellows hose).

- M-15. MDC Manufacturing, 210 Littlefield Ave., So. San Francisco, CA 94080 (vacuum hardware and flanges).
- M-16. MKS Instruments, Inc., 22-24 Third Ave., Burlington, MA 01803 (welded all-metal capacitance manometers).
- M-17. Normetex S.A., 13 Rue de la Brasserie, 27500 Pont-Audemer, France (bellows-sealed orbiting-spiral vacuum and gas transfer pumps).
- M-18. Pave Technology Co., P.O.Box 416, Vandalia, OH 45377 (hermetic electrical feedthrus).
- M-19. Nupro Co., 4800 E.345th St., Willoughby, OH 44094 (metal-bellows valves--manually and pneumatically operated).
- M-20. Perkin-Elmer Vacuum Products, 6509 Flying Cloud Dr., Eden Prairie, MN 55344 (flanges, feedthrus, ion, sorption and cryopumps).
- M-21. SAES Getters/USA Inc., 1122 E.Cheyenne Mtn. Blvd., Colorado Springs, CO 80906 (getters, getter pumps and panels).
- M-22. S.R.T.I., B.P.No.10, Route de Guyancourt, 78530 Buc, France (bellows-sealed, rolling-plate vacuum pumps).
- M-23. The Quantum Mechanics, Tracy, CA (Vacuum hardware with special surface treatments for tritium service).
- M-24. Thermionics Laboratory, Inc., 22815 Sutro St., Hayward, CA 94544 (vacuum hardware, ion and sorption pumps).
- M-25. Torr Vacuum Products, 7121 Hayvenhurst Ave., Van Nuys, CA 91406 (vacuum hardware, all metal valves, feedthrus, etc).
- M-26. Varian Vacuum Div., 611 Hansen Way, Palo Alto, CA 94303 (vacuum hardware, all-metal and polyimide-sealed valves, sorption, cryogenic and ion pumps).
- M-27. Union Carbide Corp., Linde Div., Dept. HGR, 100 Davidson Ave., Box 444, Somerset, NJ 08873 (adsorbents).
- M-28. Bi-Braze Corp., 4 Railroad Ave., Glen Head, NY 11545 (stainless steel-aluminum tube transitions).
- M-29. Arthur D. Little, Acorn Park, Cambridge, MA 02140 (orbiting-scroll gas pumps).
- M-30. Jordan Valve, Div. of Richards Industries, Inc., 3170 Wasson Rd., Cincinnati, OH 45209 (all-metal sliding-gate pressure-regulating and control valves).

- M-31. Air-Vac Engineering Co., 100 Gulf St., Milford, CT 06460 (gas-operated venturi vacuum pumps).
- M-32. Fox Valve Development Co., Inc., 2 Great Meadow Lane, East Hanover, NJ 07936 (gas-operated venturi vacuum pumps).
- M-33. Ergenics, A Div. of MPD Technology Corp., An Inco Co., 681 Lawlins Rd., Wyckoff, NJ 07481 (metal-hydride gas storage alloys and systems).
- M-34. Sargent-Welch Vacuum Products, 7300 N. Linder, Skokie, IL 60077 (turbomolecular and cryogenic vacuum pumps).
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- M-38. High Vacuum Equipment Corp., 110 Industrial Park Rd., Hingham, MA 02043 (large cryopumps).
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TRANSFER OPERATIONS WITH TRITIUM-A REVIEW

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KEYWORDS: tritium, transfer, handling

ABSTRACT

Controlled thermonuclear reactors will involve pumping operations with tritium that may involve pressures ranging from submillipascals to megapascals. A variety of pumps is available that can cover portions of this range, and these can be staged to cover the entire pressure range. Some of these pumps can be adapted to virtually any size requirement currently anticipated. Special attention must be paid to operating features and construction materials.

INTRODUCTION

Tritium is one of the most disagreeable fluids to pump because it is radioactive, highly active chemically, and very expensive. Pumping operations that exclude all contaminants without loss of tritium must be accomplished. Controlled thermonuclear reactor (CTR) applications may involve pressure ranges from submillipascals to megapascals. To cover this range, several pumping stages must be used.

Certain features, such as no moving shaft seals and no organic fluids or elastomer seals, are common to the pumps described here for tritium service. Certain vacuum pumps that do not conveniently yield tritium after pumping, such as the titanium sublimation pump, are not considered here.

For this purpose, pumps will be classified either as general-service compression pumps or as special-purpose pumps. The latter are of more limited application either because of their mode of operation or their available sizes.

COMPRESSION PUMPS

Liquid-helium cryopumps are simply surfaces chilled to 4.2 K to freeze tritium. Surfaces used require low emissivities and may include polished copper and aluminum, as well as silver-plated stainless steel. The liquid-helium reservoir is jacketed by a liquid-nitrogen chamber having a chevron or other baffle at the inlet (Fig. 1).

The ultimate pressure for these cryopumps is determined by the hydrogen isotopes present at 4.2 K, shown in Table I.¹ If a sticking coefficient of one is assumed for the cryotrap surface, the maximum pumping speed equals the conductance of an equivalent orifice.

$$S_{\max} = 3.64 A \left(\frac{T_{\text{gas}}}{M} \right)^{1/2}, \quad \text{liter/sec},$$

where

A = surface area, cm^2

T_{gas} = temperature, K

M = molecular weight.

Maximum speeds are also listed in Table I, expressed as $\text{liter}/(\text{cm}^2 \text{ sec})$. As a practical matter, pumping speeds are often limited by inlet conductances, and working pressures may range from <0.1 to 100 mPa ($\sim 10^{-8}$ to 10^{-3} Torr).

Offgassing, or compression, is achieved by warming the cryopump surfaces to $\sim 20 \text{ K}$, with resulting vapor pressures for the isotopes shown in Table II.¹ The modest temperature change needed will permit relatively rapid cycling of these cryopumps. They are well suited for large volume applications requiring high speeds, such as CTR injector arrays.

Cryosorption pumps are cryopumps that contain a surface-active material such as charcoal or

molecular sieve bonded to the cryogenic surface. Operation at temperatures in the range of 15 to 25 K will give ultimate pressures similar to those for cryopumps at 4.2 K. However, the cryosorption materials must be heated much higher to desorb fully; temperatures up to 523 K may be needed with molecular sieve.² Cryosorption pumps are more suitable for applications that do not require rapid cycling.

The *mercury vapor diffusion pump* was invented by Gaede³ in 1915. The present concept, in which the mercury vapor jet is promptly condensed, is attributed to Langmuir.⁴

A modern mercury diffusion pump is shown in Fig. 2. Mercury as a fluid is nonreactive with tritium, and thus provides good service in moderate- to high-vacuum conditions. Models presently available have speeds ranging from 2 to 10,000 liter/sec and all have ultimate pressures below 0.1 mPa (8×10^{-7} Torr). Most models will

tolerate high backing pressures, ranging up to 0.5 kPa (4 Torr).

Mercury vapor ejector pumps are multistage pumps operating at high boiler pressures to achieve high backing pressures. Ejector pumps

TABLE I

Ultimate Pressures and Maximum Pumping Speeds for Hydrogen Isotopes with Liquid-Helium Cryopumps at 4.2 K

Isotope	P, mPa (Torr)	S_{max} , liter/(cm ² sec)
H ₂	5×10^{-2} (4×10^{-7})	23
D ₂	7×10^{-7} (5×10^{-12})	16
T ₂	4×10^{-9} (3×10^{-14})	13

TABLE II

Offgassing Pressures for Hydrogen Isotopes at 20 K

Isotope	Pressure, kPa (Torr)
H ₂	90 (676)
D ₂	29 (221)
T ₂	16 (120)

Fig. 1. Liquid-helium cryopump.

Fig. 2. Mercury vapor diffusion pump.

are not true diffusion pumps, since the gas being pumped is usually in laminar or turbulent flow.

Power and Dennis⁵ have described a three-stage pump (Fig. 3), with a boiler pressure of 53 kPa (400 Torr), that can pump against a backing pressure of 13 kPa (100 Torr). This pump has an ultimate pressure of ~ 0.13 Pa (1×10^{-3} Torr) but other versions can pump to < 0.1 mPa. Ejector pumps are well suited to back up diffusion pumps since their high backing pressures can be staged to a variety of other pumps.

Fig. 3. Mercury vapor ejector pump. (Courtesy of Edwards Vacuum Components, Crawley, Sussex, England.)

TABLE III
Vapor Pressure of Mercury

$T, ^\circ\text{C}$	Pressure, Pa	(Torr)
25	0.24	(1.8×10^{-3})
0	0.024	(1.8×10^{-4})
-30	6×10^{-4}	(4.8×10^{-6})
-78	4×10^{-7}	(3×10^{-9})
-196	$< 10^{-27}$	$(< 10^{-29})$

All lines to mercury pumps must be protected against mercury vapor. This can usually be done by some kind of chilled baffle or trap. Vapor pressures for mercury at several temperatures are given in Table III.

Wobble pumps were especially developed by SRTI^a for the French nuclear industry to accommodate radioactive and other hazardous gases. Compression is achieved by a piston with a wobble motion similar to a nutating piston, shown in Figs. 4 and 5. Each side of the piston has inlet and exhaust ports without valves. The piston wobbles but does not rotate, and shaft seals are avoided by sealing the chamber with a metal bellows.

Wobble pumps have high speeds, especially at low compressions, as circulation pumps. Performance curves for three models are shown in Fig. 6, with the pumping speed plotted as a function of the ΔP , at a constant atmospheric discharge pressure. Other curves show effective pumping at inlet pressures as low as 1 to 10 Pa (8×10^{-3} to 8×10^{-2} Torr). Wobble pumps can be staged in

Fig. 4. Wobble pump cross section. (Courtesy of SRTI, Buc, France.)

^aSociété de Recherches Techniques et Industrielles, Route de Guyancourt, 78530 Buc, France. (The North American representative is Airco Temescal Division, Air Reduction, Inc., Berkeley, California. Reference to a company or product name 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.)

series to achieve higher compression, and can be used in series with diffusion or ejector pumps to give pressures up to 100 kPa (1 atm).

Diaphragm compressors use a metal diaphragm pulsed by hydraulic fluid to compress the gas, as shown in Fig. 7. Double diaphragms with a sensing port between to detect leakage can be used for tritium service.

Displacements cover a wide range up to at least 28 liter/sec. Inlet pressures can be as low as 1.3 kPa (10 Torr). Multistage pumps can provide discharge pressures well over 1 MPa (10 atm)—far beyond anticipated CTR needs for tritium service.

SPECIAL-PURPOSE PUMPS

The original *Toepler pump* was devised by Geissler in about 1855.⁶ It consisted of a vessel that was alternately filled and emptied of mercury, connected to the chamber being evacuated and to

the atmosphere by stopcocks. Toepler eliminated the stopcocks and used mercury to make all seals. A modified form is shown in Fig. 8.

Toepler pumps have been used at Lawrence Livermore Laboratory (LLL) patterned after Urry and Urry,⁷ in which automatic operation is controlled by electrical circuits that are completed through contact with the mercury reservoir, as shown in Fig. 9. With all mercury in the lower chamber, the circuit is completed through electrodes 1 and 2; compressed air introduced at A drives the mercury into the upper chamber. This seals the inlet tube at B and drives the gas through the outlet valve until mercury closes the circuit through electrodes 1 and 3. Compressed air is then shut off and vacuum from a mechanical pump is introduced at A, returning the mercury to the lower chamber and finally opening the inlet tube at B. Mercury present at both float valves seals them.

Toepler pumps have slow speeds and ultimate pressures are on the order of 1 Pa (8×10^{-3} Torr). They are now used primarily for collecting gas samples. These pumps are usually made of glass

Fig. 5. Wobble pump cutaway view. (Courtesy of SRTI, Buc, France.)

Fig. 6. Performance curves for wobble pumps at constant outlet pressure of 101.3 kPa (760 Torr). Pumping speeds are for air. (Courtesy of SRTI, Buc, France.)

Fig. 7. Diaphragm compressor. (Courtesy of Pressure Products Industries, Hatboro, Pennsylvania.)

so their use with tritium is limited to a few curies,^b at most, for reasons of safety. Toepler pumps have been used at LLL for many years with gas burettes for gas sampling with excellent results.

The *Sprengel pump* consists of a small tube through which mercury flows in a succession of drops, entraining the gas to be pumped, as shown in Fig. 10. Herman Sprengel invented the pump, which bears his name, in about 1855.⁶ Many variations have been developed, and Waran even devised a two-stage pump with automatic operation.⁸

A modern Sprengel pump is shown in Fig. 11. Mercury is moved up a central tube by a piston with a metal bellows. Pumping speeds fall sharply at low pressures, and ultimate pressures $\lesssim 1$ Pa (3×10^{-3} Torr) take a long time. Maximum pressure is generally taken as ~ 100 kPa, but the maximum pressure rise is actually determined by

^bTritium has 2.7 Ci/cm³ STP.

Fig. 8. Principle of the Toepler pump. (Sketched after *Encyclopedia Britannica*.)

the height of the falling mercury column. These pumps are useful for both circulation and transfer (compression) operations.

Sprengel pumps entrain mercury droplets in the gas being pumped. In one case, a small cyclone separator has been used successfully to remove mercury drops. Hickman⁹ has proposed a gallium-filled Sprengel pump for tritium operations to eliminate the presence of mercury vapor.

Metal bellows pumps correspond to piston compressors, except that a sealed metal bellows^c (Fig. 12) is used instead of a conventional piston and cylinder. A model incorporating secondary

^cMetal Bellows Corporation, 1075 Providence Highway, Sharon, Massachusetts 02067.

Fig. 9. Automatic Toepler pump. (Sketched after Urry and Urry.)

containment that is suitable for tritium service is shown in Fig. 13.

Inlet pressures range from 17 to 50 kPa (125 to 375 Torr), while outlet pressures reach 300 kPa (3 atm). Maximum speeds are all <3 liter/sec. Metal bellows pumps are suitable for circulation and transfer operations of limited size and/or intermittent operation.

Diaphragm metering pumps are similar to diaphragm compressors, as shown in Fig. 14. Double diaphragms with sensing ports between them are also available. There is usually an adjustable linkage to vary the length of the hydraulic piston stroke, and thus the displacement of the diaphragm. The pistons are usually geared to produce one or two strokes per second.

These pumps are of particular interest for small metered flows, such as to certain analytical instruments. For these uses, sizes are generally

Fig. 10. Principle of the Sprengel pump. (Sketched after Waran.)

<20 cm³/sec. Compression may be limited because the dead volume in the metering head is often large; however, diaphragm pumps can operate over a broad pressure range, beginning at <10 kPa (<75 Torr). Some applications may require that the flow pulses be dampened.

Cryopumping and *thermopumping* simply involve filling a container at one temperature and venting it at a higher temperature and pressure. A container filled at -196°C and warmed to room temperature would ideally increase the pressure of the gas by a factor of 3.8.

At least one company^d has made a commercial thermopump (Fig. 15). Two coils are used in series as containers. They are alternately cycled from room temperature to 315°C , giving each stage a maximum compression factor of 2; actual compressions are somewhat less to achieve a

^dPressure Products Industries, Hatboro, Pennsylvania.

Fig. 11. Multitube Sprengel pump, metal construction.

reasonable speed while expanding the gas into some final container (not shown). These pumps can be operated over the range of ~ 10 kPa (75 Torr) to >1 MPa (10 atm). For tritium service, the rupture device would have to be vented to a tank for later recovery.

CONSTRUCTION FEATURES

Special attention has recently been focused on the quality of materials used and the quality of

Fig. 12. Metal bellows pump. (Courtesy of Metal Bellows Corporation, Sharon, Massachusetts.)

welds in fabricating equipment for tritium service. Structural materials in contact with tritium, in general, should be limited to stainless steels, aluminum, or oxygen-free high-conductivity copper. Joints should be made by welding or brazing with low-cadmium silver solder. Special attention must be given to material compatibility if mercury will also be present.

Most equipment for multigram tritium service at LLL is now being made from special quality Type 316 stainless steel.¹⁰ Specified metallurgical processes are used, and particular attention is paid to chemical composition, grain size, and inclusions. Such material is literally followed from furnace to final installation. Even welding wire is specially made from this same material, and welders are specifically trained to weld equipment for tritium service.

Organic and elastomer materials will be degraded in contact with tritium, and will also contaminate the tritium by isotopic exchange. O-ring seals must be replaced with metal seals. Suitable materials include Types 304 and A286 stainless steels that have been plated with gold, silver, or lead. Soft iron gaskets with knife-edge flanges

Fig. 13. Metal bellows pump with secondary containment of the welded bellows assembly. (Courtesy of Metal Bellows Corporation, Sharon, Massachusetts.)

Fig. 14. Diaphragm metering pump. (Sketched after Pulsafeeder Products, Interpace Corporation, LeRoy, New York.)

have also been used with tritium and should be suitable in the presence of mercury.

Finally, components having a high risk of failure (e.g., metal bellows) are enclosed in a secondary container. This was shown earlier with the metal bellows pump in Fig. 13.

Fig. 15. Thermopump for operation between 25 and 315°C. (Courtesy of Pressure Products Industries, Hatboro, Pennsylvania.)

CONCLUSIONS

Transfer operations involving tritium can be accomplished by using a variety of pumps. By staging pumps, tritium can be pumped from <0.1 mPa ($\sim 10^{-6}$ Torr) to $>>1$ MPa (10 atm). These pumps can be sized to virtually any CTR or other tritium application. Close attention must be paid to the materials and methods of fabrication for tritium service.

ACKNOWLEDGMENT

This work was performed under the auspices of the U.S. Energy Research and Development Administration.

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HIGH-PRESSURE TRITIUM EQUIPMENT

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KEYWORDS: tritium

ABSTRACT

Some solutions to problems of compressing and containing tritium gas to 200 MPa at 700 K are discussed. The principal emphasis is on commercial compressors and high-pressure equipment that can be modified easily by the researcher for safe use with tritium. Experience with metal bellows and diaphragm compressors has been favorable. Selection of materials, fittings, and gauges for high-pressure tritium work also is reviewed briefly.

INTRODUCTION

Fusion-energy research is rapidly increasing the need for better tritium technology. Laser-induced fusion will require extending this technology to higher pressures and temperatures. Though several problems persist in the compression and containment of tritium under these conditions, a large backlog of experience already is available. Los Alamos Scientific Laboratory (LASL) has a great deal of experience handling tritium gas at pressures as high as 175 MPa (25,000 psi)^a and at temperatures to 700 K. At these conditions, it is possible to permeate hollow laser-fusion targets with deuterium-tritium (DT) gas in relatively short times.

The purpose of this presentation is to review some of the problems of handling tritium gas at high pressures and temperatures and the approaches we have taken toward their solution.

Most of the remaining problems could be solved by today's technology if tritium users could clearly communicate their needs to the manufacturers.

GENERAL CONSIDERATIONS

The problems of containment of tritium at high pressures are essentially the same as for hydrogen. All structural members must be relatively impermeable to hydrogen and absolutely free from hydrogen embrittlement. These requirements are made more stringent by the fact that tritium is radioactive, toxic, and expensive. Austenitic stainless steel generally provides adequate strength at both cryogenic and ambient temperatures. Where higher strength or lower bulk is required, composite vessels can be built with austenitic stainless steel as the liner. For high-temperature autoclaves, stainless steels are unsatisfactory because of the high hydrogen permeabilities of iron and nickel. We have used beryllium copper to 675 K with some success. Other approaches are to use a composite vessel with a low permeability liner (copper or aluminum) and a high-strength shell, or a cold-wall vessel with the insulated, heated reactor immersed in pressurized gas.¹

Any tritium system built today must meet extremely low emission standards; secondary containment is the only reliable means of staying below such release levels. For a high-pressure system, this can present serious design problems. The secondary container must enable recovery of the tritium released by a failure, and must withstand damage from high-velocity fragments ejected at the same time. It is desirable to design high-pressure components for a negligible probability of catastrophic failure and to rely on the containment system to control only leakage.

Another problem introduced by tritium involves

^aOne megaPascal (MPa) is equal to 145.0 lb/in.².

the choice of seal materials—compression gas-kets, valve packings and seats, etc. The average tritium beta particle (6 keV) has several orders of magnitude more energy than any organic bond. Tritium also is active chemically and exchanges readily with normal hydrogen, accelerating the decomposition of organic materials. Slow deterioration of plastics and elastomers can be tolerated perhaps in a vacuum or atmospheric pressure system, but pure tritium at 200 MPa generates radiation at a density approaching 2000 Ci/cm³; the useful life of all elastomers and most plastics can be measured in hours. It is often overlooked that the decomposition products of some organic materials are also highly corrosive. The fluorocarbons, which decompose into tritium fluoride, are a particularly vicious example. We once traced an epidemic of metal-bellows failures in a low-pressure system to this cause.

The solution is to use metal gaskets wherever possible and to design any necessary plastic seals so that a minimum surface is exposed to the tritium. Where the resiliency or low friction of a plastic is still required, as in high-pressure valve packings or soft-seat check valves, we have found the polyimides² to be the most radiation resistant. They also have satisfactory physical properties for high-pressure seals.

VALVES AND FITTINGS

Most commercial high-pressure fittings are intrinsically suitable for use with tritium, but users are advised to set up their own certification and acceptance standards to supplement the quality-control procedures of the manufacturer. This is only prudent when all the costs of an accidental release of tritium are considered. The fluorocarbon stem packings in commercial high-pressure valves can be successfully replaced with polyimide plastic. This not only improves the radiation resistance, but also reduces packing extrusion and cold flow. At LASL, we use a rotary pneumatic actuator³ as a remote valve operator (Fig. 1). We prefer this device to the diaphragm-type operator available from most manufacturers because the rotary design limits both stem travel and closing torque. Thus, valve seats and packings seldom require replacement, and infrequent maintenance is the most important factor in reducing personnel exposures.

GAUGES

Gauging is a particularly demanding problem because tritium is a controlled radioactive material, and its accountability at high pressure is best done by accurate pressure-volume-tempera-

Fig. 1. Remote high-pressure valve operator.

ture measurements. Gauges that operate by deflection of an elastic member necessarily incorporate smaller safety factors than desired for conservative design. Nevertheless, Bourdon-tube gauges have served the pressure industry for more than a century, and are a good choice for tritium if the Bourdon material is hydrogen-compatible. Many manufacturers furnish Bourdon gauges in austenitic stainless steels or beryllium copper in a wide range of pressures. The addition of force-balancing and digital-readout techniques has greatly enhanced the precision of these inherently simple devices. Strain-gauge transducers are preferred by many experimenters now. Solid-state digital electronics and improved quality control have resulted in precisions rivaling those of the force-balance Bourdon-tube systems. All-welded transducers of tritium-compatible materials are also available from several manufacturers.

COMPRESSORS

The principal constraints on tritium pumps, enumerated by Folkers and Gede,⁴ are that they incorporate no moving shaft seals, elastomers, or organic fluids. Compressors that readily can meet these requirements generally fall into one of three classes: (a) mercury-column compressors, (b) thermal-cycle and change-of-phase pumps, and (c) flexible metal-membrane compressors.

Fig. 2. Mercury-column compressor.

Mercury is compatible with hydrogen and was used in many early tritium compressor designs⁵ because of the ease with which shaft seals could be protected by liquid metal. Although mercury is not attacked by tritium, it is incompatible with other common pressure-system materials, and effective trapping of vapor and droplets carried by a dense, high-velocity gas is difficult. If the pressure system contains no amalgamable metals, and if some level of mercury vapor can be tolerated in the compressed tritium, a commercial mercury-column compressor⁶ may provide a ready solution (Fig. 2).

Classification (b) includes plain and adsorbent-assisted cryopumps, hot-gas thermal compressors, and metal-hydride pumps.⁷ Devices of this type are simple in design and contain no moving parts. Only static seals are required for closing the working volume and introducing any electrical leads; many satisfactory solutions have been developed for these. The main limitations on these designs are: limited capacity in a reasonable physical size and slow cycle speed. Small volumes can be pumped to quite high pressures with one of these devices of modest dimensions if the experimenter can wait an hour or so between cycles. These pumps are not available from com-

mercial sources as complete systems, but often can be assembled from commercial high-pressure components.

Classification (c) comprises compressors that employ metal bellows or diaphragms. Several manufacturers make diaphragm compressors.^{6,8,9} The best models for tritium incorporate a stainless-steel diaphragm welded to the gas head. Since reliable welds are difficult to make for service above 100 MPa, we are using a hydraulically loaded seal of gold plating on the gas diaphragm. At 140 MPa, no tritium leakage can be detected with the usual portable air monitor, and diaphragm life is almost infinite if the gas is free of particulates. The utilization of triple diaphragms⁸ (Fig. 3) with an appropriate vent port can give effective secondary containment for the most likely mode of compressor failure. At LASL, we have used a multistage diaphragm pump routinely to compress tritium to 140 MPa. The only continuing maintenance has been frequent changes of interstage check valves, which originally had fluorocarbon face seals and elastomer poppets. These materials have now been replaced with polyimides. Some of these valves are still operating well after two years of intermittent tritium service.

Metal-bellows compressors¹⁰ have come on the market in the past few years. A design limitation of the standard compressor for tritium is excessive use of fluorocarbon plastic in the check-valve header. We have replaced this with an all-metal valve assembly. The manufacturer will provide an all-welded assembly at considerable extra cost. Because the mechanically driven bellows is unsupported, the output pressure is limited to ~0.5 MPa. The only interest in this pump for the high-pressure experimenter, therefore, is as a transfer pump or as the first stage of a multistage system.

Fig. 3. Triple-diaphragm construction.

Fig. 4. High-pressure bellows compressor.

At LASL, a balanced-pressure hydraulically driven metal-bellows compressor has been built (Fig. 4). It is designed to give a 50/1 compression ratio at 200-MPa discharge. Coupled to a suitable output volume, it can provide very sensitive pressure control by using a hydraulic proportioning pump. Driven by a conventional hydraulic plunger pump, it is capable of one 250-cm³ stroke/min.

CONCLUSION

Although several manufacturers make compressors and hardware that can be modified for

tritium use, no current item of off-the-shelf equipment is completely satisfactory. For example, every diaphragm-pump supplier uses some fluorocarbon or elastomer in the compressor even though these could be avoided. Tritium users in the fusion-energy program must fully inform industry on the requirements for tritium handling. Now that tritium is emerging as a vital fuel for the world's future energy needs, we can and must apply the best current technology to its safe handling and containment.

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A TRITIUM-COMPATIBLE HIGH-VACUUM PUMPING SYSTEM

by

Don O. Coffin

I. INTRODUCTION

Since tritium was first produced in nuclear reactors, users have sought ways to contain and handle this very active radioisotope safely. Now the prospect of success with fusion power reactors challenges vacuum engineers to design high-vacuum systems capable of pumping large quantities of tritium gas. All magnetic confinement reactors—tokamaks, mirror machines, and bumpy toruses—require high vacuums ($10\ \mu\text{Pa}$) and pumping speeds ($100\ \text{m}^3/\text{s}$) for mixtures of hydrogen and helium isotopes. To these already overwhelming performance requirements are added the complications of handling tritium, with all its compatibility and radiogenic problems.

Compound cryopumps¹⁻⁴ will evacuate hydrogen/helium mixtures from plasma chambers at speeds and to base pressures required. Cryopumps scale up readily and do not present any insuperable tritium-compatibility problems. Some of the plasma machines currently under construction will employ wall pumping in the form of nonevaporable getter panels mounted inside the plasma chamber.⁵ These devices also can be scaled up by simply increasing the panel area, and they are safe and effective for tritium.⁶ Although the speed, high vacuum, and tritium-compatibility requirements can be met with compound cryopumps, getter pumps, or a combination, removing the expended deuterium-tritium (DT) mixture from the plasma chamber is only the first step in a complex process of recovering and purifying the fuel for reuse. Cryopumps and getter panels are reservoir devices, which must be regenerated periodically before their capacities are exceeded. The exigencies of this regeneration process define the requirements of a vacuum pumping system as follows:

- No impurity contamination of the gases pumped;
- No deterioration of the pump materials by tritium;
- All mechanisms hermetically sealed;
- No holdup or inventory of tritium in the pump train;

- Base vacuum at the pump inlet below $1\ \text{mPa}$;
- Ducted, completely contained pump exhaust at $100\ \text{kPa}$.

The first two requirements preclude any organics or fluorocarbons (elastomers, plastics, or lubricants) in pump chambers, where these materials can interact with the tritium-rich vacuum medium.

The need for a pumping system meeting these requirements has long been recognized, even predating consideration of vacuum systems for fusion reactors, but no pump combination meeting all of these requirements has been available. This paper reviews attempts to solve these problems and describes a pumping system assembled at the Los Alamos Tritium Systems Test Assembly (TSTA), which completely satisfies the special requirements of fusion vacuum systems.

II. FOREPUMPS

A. Liquid-Sealed Pumps

Gas transfer pumps operating in the viscous pressure regime have been available for many years,^{7,8} and tritium has been pumped with nearly every type of mechanical pump available. To avoid problems of compatibility with rotary pump oils, tritium users resorted to various mechanical pumps with a mercury sealant and pumping medium. Both Toepler⁹ and Sprengel¹⁰ pumps have been used successfully for tritium transfer. The main difficulties with mercury are its mobility and its propensity to amalgamate with all sorts of vacuum materials. Cryogenic trapping and fast-closing automatic valves are required to confine the mercury to the pump, but even with elaborate precautions, mercury usually finds its way to precious metal or copper components of the vacuum system. The vacuum designer usually discovers that mercury is as difficult to control and contain as is tritium itself.

B. Dry Scroll Pumps

One recent development is the scroll pump.¹¹ The pumping action of this device will be described subsequently under the discussion of booster pumps. In the version suitable for the viscous pressure regime, gas is compressed between two interleaved plates with spiral channels. The plates rub during operation, so they are fabricated from graphite to reduce internal friction. This graphite scroll pump introduces shaft rotation into the pump chamber by a canned magnetic drive mechanism and converts this into the required orbital motion by an internal Oldham's coupling. This coupling also generates friction, which is minimized by fabricating those parts of polyimide. Although not ideal for tritium (both graphite and polyimide retain water and chemically react with tritium to produce tritiated hydrocarbons^{12,13}), this pump is nevertheless hermetically sealed and dry. Several models are available for gas transfer in the rough vacuum range.

C. Metal-Membrane Pumps

The metal-membrane pump is very adaptable to tritium transfer. Both metal-diaphragm and metal-bellows pumps are available from commercial sources and have been used extensively at Department of Energy (DOE) laboratories.¹⁴ At TSTA we selected a commercial metal-bellows pump* for general gas transfer throughout the tritium circulation and purification loop. The Los Alamos version is 100% hermetic with metal static seals and all-metal check valves (Fig. 1). With multiple staging this pump can compress to 300 kPa and evacuate to 2 kPa. It is our choice for the final-stage forepump of a tritium-compatible vacuum pump system.

III. HIGH VACUUM PUMPS

A. Diffusion Pumps

In the molecular flow regime (below 1 Pa) there are several possibilities for pumping tritium gas mixtures. The oldest are mercury diffusion pumps, which have been used extensively for tritium, especially at facilities where Toepler or Sprengel pumps are used for transfer

*MetBel Model MB-601, Metal Bellows Corp., 1075 Providence Hwy, Sharon, MA 02067.

Fig. 1.
Reciprocating metal-bellows pump.

operations at higher pressures. Mercury compatibility problems preclude this class of pumps for fusion vacuum systems.

B. Capture Pumps

Ion and titanium-sublimation pumps work well with hydrogen isotopes to very low pressures, but because these devices bury pumped gases, for the most part irreversibly, tritium cannot be completely recovered, so they do not meet the zero-inventory requirement. Such pumps are therefore unsuitable for regenerating large plasma-chamber cryopumps. Cryopumps and non-evaporable getter pumps were discussed in connection with plasma chamber pumping. Although these devices efficiently evacuate hydrogen (getter pumps cannot pump helium), the regeneration problem is only postponed by using one cryopump to regenerate another.

C. Turbomolecular Pumps

There have been exciting developments in turbomolecular pump technology in recent years. Early turbopumps did not have adequate compressions nor speeds for light gases. Moreover, tritium compatibility was a problem because of the organic lubrication required for the high-speed bearings exposed to the

vacuum medium. Failure of the turbopump power supply could result in contamination of the high-vacuum chamber from diffusion of oil vapor. Turbopumps that overcome these difficulties now are manufactured.

One new model has gas bearings* and can be driven by either a conventional frequency converter or an air motor. The disadvantage of this pump for fusion applications is its low pumping speed for hydrogen and helium. Moreover, part of the bearing gas (air or nitrogen) becomes admixed with the exhaust going to the forepump, giving large amounts of high-mass impurities that must be subsequently separated from the DT. Another design** suspends the high-speed rotor in vacuum with a magnetic field, thus eliminating mechanical bearings and their lubrication. This magnetic-bearing turbopump (Fig. 2) is our choice for the first or high vacuum stage of the tritium-compatible vacuum system. It has a speed in excess of 500 L/s for both hydrogen and helium and tolerates forepressures as high as 10 Pa. As supplied by the manufacturer, this pump contains a few elastometric static seals, but these are replaced easily with metal wire rings to achieve complete tritium compatibility.

*Alcatel Model 5450, Alcatel Vacuum Prod., 60 Sharp St., Hingham, MA 02043.

**Leybold Model TMP-560M, Leybold-Heraeus Vacuum Prod., 5700 Mellon Road, Export, PA 15632.

Fig. 2.
Magnetic-bearing turbomolecular pump.

IV. BOOSTER PUMPS

The pressure regime between a few kPa and a few Pa has always been the most difficult for tritium pumping. All high-vacuum pumps suitable for tritium require a forepressure below a few Pa to operate. (The Alcatel* gas-bearing turbopump can maintain a high vacuum without a forepump, but the high vacuum must be established by other means initially). Because the metal-bellows forepump cannot generate vacuums below 2 kPa, we require some type of booster pump to span the transition pressure range.

A. Roots Blowers

Roots blowers show promise for tritium in the transition pressure regime because a number of canned-motor Roots machines are available. Even though these pumps can be hermetically sealed, tritium-compatibility problems remain because of lubrication required for the gears and bearings that synchronize the high-speed, controlled-clearance rotating parts. There is one report¹⁵ of a Roots pump with Ferrofluid† shaft seals. Only one of these devices was ever built, and its speed and compression ratio were inadequate for our requirements.

B. Rolling Plate Pumps

Another gas circulating pump of considerable interest is the "wabble" (or PR for plateau rotulant, or rolling plate) machine manufactured in France.†† This machine uses a conical-faced disk, nutating in a close-fitting chamber. The nutation is introduced into the vacuum chamber by a bent shaft sealed by a rocking metal bellows. The PR pump exhibits high speeds in the transition flow range and also has the required hermetic drive. Unfortunately, compressions are generally limited to less than 10 to 1 per stage. Therefore, several pumps in series are needed to bridge the pressure range from 2 kPa (metal-bellows inlet) to the 10 Pa required to back a turbopump.

†Ferrofluidics Corp., 40 Simon St., Nashua, NH 03061.

††SRTI Model PR, Soc. de Recherches Techniques et Industrielles, Rte. de Guyancourt, BP No. 10, Buc, France.

C. Moving Spiral Pumps

The most promising vacuum boosting device for tritium is another French pump¹⁶ that also uses a nutating bellows to seal the motion of the compression member. The gas compression geometry (Fig. 3) is a thin-walled cylinder, the generator of which describes an involute, or spiral of Archimedes ($R = A \cdot \theta$). The pump cavity is enclosed between two of these clock-spring spirals: one fixed to the pump base by a rigid cylindrical barrel; the second generated 180° out of phase to the first and displaced from it by a vector whose magnitude is the pitch of the spiral. The pumping action is provided by uniformly rotating this displacement vector which results in a small orbital motion of one spiral within the other. An inlet port is located near the outer edge of the stationary spiral and the discharge port at its center. The progressive positions of the moving spiral (Fig. 4) show how the orbital action first traps, and then continuously compresses the gas until it is discharged through the center port.

The spiral plates are fabricated using numerical-control machine tools and are hand-lapped at assembly, so that clearances of about 10 μm can be maintained at all times between the fixed and moving spirals. The moving parts never contact, so internal friction is eliminated, and high orbital frequencies (60 Hz) are possible. The design permits multiple stages for high compressions, because each full turn on the spiral pair creates an independent trapped gas volume. Thus both the input displacement and overall compression are increased by adding turns to the spiral, and several successive compression stages can be machined into the single moving plate.

Fig. 4.
Orbital positions of spiral booster pump.

The hermetic drive is revealed in a vertical section through the pump (Fig. 5). A long hydroformed metal bellows is welded to the bottom periphery of the moving plate and to the stationary pump base. The orbiting plate moves the upper end of the bellows in a circular path, but there is never any component of motion that introduces torsion between the two ends of the bellows. Because it experiences only a small periodic axial deflection, the bellows should have a very long life in this application. A

Fig. 3.
Moving-spiral vacuum booster pump.

Fig. 5.
Spiral pump drive and sealing mechanism.

motor-driven crank generates the orbital motion at one vertex of an equilateral triangle near the outer edge of the moving plate. Two additional idler cranks constrain the plate motion at the other two vertices. The location of all eccentrics outside the vacuum-sealing bellows permits them to be fitted with heavy-duty bearings with conventional lubrication. This three-point peripheral drive provides the precise orbital motion required for this high-speed, small-clearance pumping mechanism.

Although this pump is the only hermetic tritium-compatible booster device that offers high compressions (in excess of 10,000 to 1 at blank-off) in the transition flow regime, its use is not limited to pressures below 2 kPa. A 4-L/s spiral pump tested at Los Alamos performs well at inlet pressures as high as 10 kPa if its discharge port is maintained below 15 kPa by an auxiliary forepump. This range is ideal for metal-bellows compressors, which develop maximum performance between 5 and 200 kPa. Larger pumps (40-160 L/s) of this design have better volumetric efficiencies that allow them to exhaust directly to atmospheric pressure (100 kPa). Such pumps have been pumping UF_6 continuously for several years in Europe and elsewhere. A 5-L/s pump with aluminum spirals (for better heat dissipation) is now in production,* and it too will offer high compression ratios without requiring a forepump. The measured

performance of a prototype of this pump is given in Fig. 6.²²

At blankoff, the 4-L/s pump tested at Los Alamos achieved ultimate vacuums of 0.5 Pa with two stages of metal-bellows forepumping (7 kPa at the spiral pump discharge port). When another stage of metal-bellows compression was added (reducing the forepressure to 2 kPa), the spiral pump inlet pressure dropped to less than 0.1 Pa. These blankoff pressures in the high ion-gage range were achieved in less than 1 h with no bakeout nor cryogenic trapping.

Fig. 6.
Spiral pump performance.

*Normetex S.A., 13 Rue de la Brasserie, 27500 Pont-Audemer, France.

The pump spirals are designed so that both adiabatic heating and compressional deflections tend to increase clearance between the fixed and moving plates. Although this reduces efficiency, the pump mechanism is not endangered by exposing the inlet to atmospheric pressure at full speed. This feature eliminates the need for a bypass during initial pumpdown, and if the spiral pump is turned on at the same time as the metal-bellows fore-pump, it makes a significant contribution to the pump-down even before its optimum fore-pressure is reached.

Roots blowers generally exhibit poorer performance for helium and hydrogen than for the heavier gases because of backleakage past the rotors during compression, and carry-back of gas molecules adhering to the rotor as it returns to the high-vacuum side of the chamber. Both effects are more pronounced for the more mobile light gases. There is no carry-back effect in the spiral pump because it has no rotor; and because spiral pump clearances are smaller than for Roots blowers, backleakage is also less. Most of our tests on the 4-L/s spiral pump have been run with helium or deuterium, and we have consistently observed higher pump speeds than manufacturer's specifications for air. In fact we observe no difference in the pump's performance with air or helium above 100 Pa, an important feature for fusion vacuum systems.

V. THE TSTA CRYOPUMP REGENERATION TRAIN

A. Components

At the Los Alamos TSTA we have assembled a series of hermetic, tritium-compatible mechanical pumps, chosen from those described in the previous sections. This pumping train has been used for regenerating gases from each of the TSTA cryopumps² during preliminary testing, and it will be installed permanently in the TSTA facility for regenerating deuterium-tritium-helium from the cryopumps when TSTA becomes operational in 1982. A 500-L/s magnetic-bearing turbopump, a 4-L/s moving spiral pump, and a two-stage metal-bellows pump are connected in series for the TSTA application (Fig. 7). Running the frictionless turbopump continuously is advantageous, so valves are provided for its isolation during initial pumpdown and up-to-atmosphere operations. The turbopump has safety interlocks, which

TYPICAL PRESSURE GAGE READINGS (Pa) AT BLANK-OFF

Fig. 7.

A tritium-compatible vacuum pumping system.

cut the power and allow a safe spindown if pressures become too high while it is isolated from the forepump system.

B. Secondary Containment

Secondary containment for tritium at TSTA is of two types. Gas is processed at pressures of 1 kPa or higher by equipment enclosed in inert-atmosphere gloveboxes. Large high-vacuum components (cryopumps, simulated plasma chamber, and primary high-vacuum ducting) are enclosed in a separately ventilated room, from which tritium can be removed by a room cleanup system in the event of a release. The metal-bellows and moving spiral pumps are located in the glovebox. The turbopump, which must be coupled directly to the cryopump if its high speed is not to be degraded by duct resistance, is located in the ventilated cryopump enclosure. The 4-cm-diam foreline leading from the turbopump to the glovebox penetration is shrouded for secondary protection. Fig. 8 is a photograph of the TSTA vacuum pump assembly before construction of the cryopump enclosure.

Fig. 8.
Assembled TSTA vacuum system.

C. Functions

Although we developed the mechanical pump train only to regenerate cryopump gases, its excellent performance and simplicity enable us to use it for all normal vacuum functions. Because the fore-pump/booster-pump combination can be used up to atmospheric inlet pressure, this system will be used for initial evacuation of the simulated plasma chamber and cryopumps. These completely dry pumps are never a source of hydrocarbon backstreaming, and vapor traps and all their attendant disadvantages are completely eliminated. Other functions that can be performed efficiently, with or without the turbopump, are:

- Recovery of DT from non-evaporable getter pumps;
- Recovery of DT from uranium storage beds;

- Pre-maintenance DT recovery and evacuation of adsorption beds, metal-getter beds, and other fuel purification components;
- Evacuation of tritium supply containers.

D. Performance

The TSTA mechanical pump train was used repeatedly to rough out each of three TSTA prototype cryopumps before and between tests. Typical pumpdown curves for air and helium are plotted in Fig. 9. The volume evacuated was approximately 0.4 m^3 and had a high surface-to-volume ratio typical of multipanel cryopumps.

Fig. 9.
Pumpdown curves for the TSTA vacuum system.

When compared to one evacuation station in those systems required for the first generation of DT-burning tokamaks,¹⁷ the TSTA cryopump system is approximately quarter-scale in pumping speed and cryopanel capacity. The mechanical pump system described here can regenerate the TSTA cryopumps, while maintaining separation of helium and DT, in about 4 h, a period comparable to the pump duty cycle. If no gas separation is attempted, the cryopump can be warmed up quickly, and regeneration times are limited only by thermal considerations.

E. Scaling Up

For the larger cryopump systems of the future, the TSTA regeneration system will scale up readily. Although pure magnetic-bearing turbopumps are now manufactured in only one size, a 6-m³/s turbopump with magnetic upper and conventional lower bearings is available.* Conventional turbopumps as large as 10 m³/s have been built. There is no fundamental reason that magnetic-bearings or some hybrid technology cannot be extended to these 10-m³/s turbos. We require only a small moving spiral pump at TSTA, but machines as large as 160 L/s are already in service elsewhere. The metal-bellows pump we are using is the largest one currently mass produced, but it is physically a very small device; building larger ones, if required, should not be difficult. Larger bellows pumps may not be required because the larger spiral pumps perform satisfactorily at atmospheric pressure discharge without a forepump. By series-staging spiral pumps, vacuums below 1 Pa can be attained without a turbopump. This is adequate for many secondary fusion applications, including mixed-gas cryopump regeneration.

VI. SUMMARY

For regenerating tritium gas from cryopumps at the Los Alamos TSTA, we have assembled a mechanical pump system that satisfies our performance requirements while remaining completely compatible with tritium and other radioactivity. It does not rely on cryogenics, chemical pumping, mercury or so-called radiation-resistant lubricants. All components of this

system are obtainable from commercial sources, and some are already available in both smaller and larger models. These pumps should have wide-spread application in fusion facilities and other places where radioactive or high-purity gases must be pumped continuously, reliably, and without contamination.

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NOTE: The moving-spiral pumps described and illustrated in this paper (pages 4-5) are built and sold under license of World Patents issued to P. Vulliez.