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1-A

TITLE (INDICATE SUBJECT CONTENT)

Tritium Calculations

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Chemical Processing Division

PERIOD COVERED (INCLUSIVE - MONTH, DAY, YEAR)

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Asmt: Investigate methods of reducing the quantity of tritium (^3H) released to the Environ. from the PUREX plant.

I. Begin Asmt:

A. Become familiar w/ PUREX flowsheet.

1. Particular atten. to paths of process condensate of ^3H .
2. Van and Ken for aid in detn. of ^3H paths and changes in ^3H paths due to planned changes in recycling condensate.

B. ~~Mr.~~ M. J. Szulinski (& others) can assist.
Bob Kiehl
Jack Kendall.

II. Liter. Search:

A. Gather info. about various meth.s or processes which might be used to isolate ^3H $\Rightarrow$ agreement or choice of those which warrant further study.

B. For selected processes:

1. Define curve relating # of stages req'd to achieve various storage vol.s / unit of product production (64 Flows) in plant.
2. Estimate capital, utilities, essential mat'l's costs.
3. Expected to document study results.

III. Monthly Summary: due once a month around the 15th.

(Include brief outline of work plan to undertake next.)

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Attack on problem: ^3H Trapping

Wild ideas: may be too far out

Freezing condensate & sep. ^3H by (?) cryogenics

Condensing slowly - in stages to get only tritiated water

Fractional Distillation for separation

Filtration

Absorption

Absorption

Decomposition $\rightarrow ^3\text{H}(\text{gas}) \rightarrow$ liquify

Remove before released into soln. - (in decont or dissoln.)

Put into "solid state" $\rightarrow$ "glased" to be stored.

Put into a chemical to be used or stored $\{2\text{r}^3\text{H}, \text{Na}^3\text{H}, \text{etc.}\}$

* side prodn. $\rightarrow$ heavy water

Questions -

✓ Decay scheme of tritium & tritiated water

where ^3H comes from - where goes (in plant process flow)

Ref: Report CEA-R 3350

7/7/70

"Means of Detecting & protecting Against Tritium"

Yves Sutra - Fourcade (Dec. 1967)

(E: 18.6 keV, max 15.6 keV, avg)

Decay: $^3\text{H} \xrightarrow[\beta^-]{100\%} ^3\text{He}$

Radioactive period: 12.262 yrs = 4520 days

Specific Activity: ^3H gas: ($^3\text{H}_2$, T_2) 2.63 Ci/cm³ or 9.7 (10³) Ci/g.

HT

1.31

"

water vapor:

T_2O

2.63

"

HTO

1.31

"

water:

T_2O

2,630 Ci/g

HTO

1,450

"

DTO

1,390

"

1 Ci (^3H) = 1.03×10^{-4} g. of $^3\text{H} \Rightarrow 9.7 \times 10^3$ Ci/g. ^3H

MPC:

2×10^{-7} $\mu\text{Ci}/\text{ml}$ in air

3×10^{-3} $\mu\text{Ci}/\text{ml}$ in water

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Nat'l ^3H : $4 \times 10^{15} \text{ } ^3\text{H}$ for atmospheric H
 $3-8 \times 10^{18}$ " fresh surf. waters
 1×10^{18} " oceanic " "

$1 \text{ } ^3\text{H} \text{ atoms} / 10^{18} \text{ } ^1\text{H} \text{ atoms} \approx 1 \text{ Tritium Ratio}$

For water @ 25°C , 1 tritium ratio = $67,000 \text{ } ^3\text{H} \text{ atoms/ml} = 3.2 \times 10^{-9} \mu\text{Ci/ml}$

max. permissible body burden for ^3H (in tritiated water) = $1 \text{ mCi} = 1000 \mu\text{Ci}$

Biological $T_{1/2}$ (in man) ≈ 10 days.

Ken H. Henry

I. possible Techniques for ^3H Containment

- A. Fuel out-gassing (pyrochem. or mech. head-end not practical w/ metal fuels - only good w/ oxide fuels)
- B. Dehydration of Dissolver Soln. (Not feasible - too "hot")
also prob. of denitration
- C. Depletion by Evaporation

II. Tritium Path Through PUREX plant

INPUT: 100% - irradiated fuels

OUTPUT: 82.9% process condensates
 15.2% + Atmosphere Dischrgs
 1.9% Hi-level Waste
 0.04% Organic Wash Wastes
 0.001% U product Soln.

+ Atmospheric ^3H dischrg obtained by mat'l bal. & not meas'd. Believed that ^3H input value, based on fiss. yield calcs., is accurate only to $\pm 20\%$. published data based on actual ^3H meas. in atm. dischrg indicate only $\sim 1\%$ of input ^3H lost via this route.

Ken has meas'd T content of dissolver off-gas, vent sys. off gas, and solvent extraction feed & waste streams.

Calcn:

Process condensate stream: $\sim 98\%$ of T (found to leave plant)

Hi-level aqueous waste stream: bulk of remaining 2%

(Calc. disregards 15% atmos. dischrg.)

Assumption:

Tritium present in solvent extraction feed soln. as tritiated water & partitions just as

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RPS (Tritium in absence of other radio. contaminants)

MPL $\approx 1. \times 10^{-3}$ $\mu\text{Ci/cc}$ for drinking water.

Non-aqueous processes: fluid-bed fluoride volatility,
pyrochem. process.

$T \xrightarrow[\text{desorption}]{\text{NaF}}$ $\xrightarrow[\text{TF}]{\text{HF}}$ activated alumina or Soda Lime $\xrightarrow{\text{water}}$ activated alumina.

J. Alkamine

7/13/70

Fractional Distillation:

Rich Pease calc'd 150 ideal stages for 10-fold conc. of deuterium (tritium out the bottom with the deuterium) which would present better results for tritiated water, but only ~50% eff. {suggests buying or writing own computer code for column}. $P = 760 \text{ mm Hg}$

Look-up processes for concentrating deuterium in water mixt. (not find much on tritium) — processes for separating water isotopes [Heavy-water plants use H_2S rxn.]

Diffusion: may be difficult since have 18 isotopes to separate but U^{235}F_6 and U^{238}F_6 have been sep'd.

Recycling tritiated water & bleeding off and storing a continuous stream after a build-up has occurred and continues ["hot" — got to shield or compensate somehow].

? How much actually goes up stack.
Need to know — if $> 1-2\%$ may have to recover.

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PUREX tech. manual

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Sat'd soln. uranyl nitrate (UN) @ 25°C ~ 72 wt. %

$UO_2(NO_3)_2 \cdot 6H_2O$ (UNH) or 2.6 moles/liter.

presence of nitric acid markedly reduces solubility of UN in water.

Fig. IV-6: Qg. U soln.

UNH (2.0 M) → 62 wt. % UNH → 48 wt. % UN
 2.6 M → 72 " " → 57 " "
 2.5 M → 71 " " → 55.5 " "

Fig. IV-11: Boiling pt. isotherms

UN (2.0 M) = 48 wt. %
 HNO_3 (0.48 M) = 1.8 wt. %
 water = 50.2 % } B.P. ~ 106.6 °C

Dissolver

stock UNH

Temp > 50°C

sp gr 1.163

UNH 2.0 M = 100.4 g

HNO_3 0.48 = 30.2 g

$P_u = f_p$

$\therefore H_2O = \frac{100.4}{33.1} = 3.03$
 596 g

B.P. ~ 106°C

$\rho_{BP} \sim 1.57$ g/ml

? H_2O
 Acid

stock UNH

Acid
 water (w/T)
 UNH (2.0 M)

Acid
 UNH (2.5 M)
 water

EVAPORATION

T°C?

How separate? % H_2O
 % HNO_3

from 101° → 120° : water rapidly removed (w/ some acid)
 @ 120°C : water + HNO_3 form max-boiling azeotrope
 and 68 wt. % HNO_3 distilled from soln.

BP of 100% UNH = 118.6° ± 0.4°C @ 760 mm Hg.

G. Mannine

7/21/70

BASIS: 1 Liter UNH soln. in @ B.P. of 106°C (2.0M UNH).

SOLN.: 1 l. soln. @ 106°C = 1510 gr.
 (753.6g) 2 M UN = 48 wt. %
INITIAL: (28.3) 0.48 HNO₃ = 1.8 "
 (788.1) H₂O = 50.2 "
 FP + PU
 sp. gr. = 1.57

(looking @ PUREX tech. Man. Fig. IV = 11?)

B.P. ≈ 108°C

2.5 M UN = 55.5 wt %

FP + PU

HNO₃ = 2.0 wt %

H₂O = 42.5 wt %

sp gr ≈ 1.71

FINAL:

$$\text{UN} = \frac{753.6}{X} = 0.555 \Rightarrow X = 1357.8 \text{ gr. in final soln.}$$

HNO₃ % 27.2
 Water % 577.0

Assumed: A 4:1 boil-off of water & tritium

No 4N evaporated over

Δ acid = 28.3 - 27.2 = 1.1 gr. } CONDENSATE
 Water = 788.1 - 577.0 = 211.1 gr }

(100%) $\frac{211.1}{788.1} = 26.8\%$ water in condensate } 1-stage evaporation.
 73.2% " " soln.

J. Akamine

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(PUREX flow sheet)

3 Streams (Tritium numbers based on

K.H. Henry's "Tritium path" calcs)

	Moles	mole fr	Sp gr	105	
HNO ₃	1.63	0.03	HNO ₃	1.63 M	= 102.7 gr./L (H ₂ O = 947.3 gr.)
H ₂ O	52.63	0.97	Pu, U	Trace	~0
	54.26	1.00	Flow	31.6	
			conc. Tritium	~ 0.619 / flow

Pu	34.5	conc. Tritium	~ 0.645 / flow
Temp	~ 52°C		
Sp gr	~ 1.0		
HNO ₃	~ 0.08 M

Flow	27.6	conc. Tritium	~ 1.75 / flow
Temp	> 50°C		
Sp gr	1.63		
U	2.0 M		
HNO ₃	~ 0.08 M		
Pu + FP			

Jean Alami

7/28/70

PROCESS CONDENSATES (CRIBS)

* AAD calc. (cont.)

Energy = $(188.7 \text{ g-moles/min.})(68.2 \text{ kcal/g-mole}) = 128,705 \text{ kcal/min.}$

conversion factor = $(4.319) \text{ (KW)} = 4 \text{ kg-cal./min.}$

$\therefore \text{Energy} = 29.8 (10^3) \text{ KW} \approx 30 \text{ MW (th)}$ energy reqmts. to handle $\sim 10 \text{ gpm. water}$ to electrolyze

J. Akaevine

7/28/70

Method of total reflux (perry 13-28)

$$n \log \alpha = \log \left[\frac{(x_1/x_2)_{\text{upper plate}}}{(x_1/x_2)_{\text{lower plate}}} \right]$$

↑
theor. stages for separation

↑
volatility sep.

1 = lighter element.

Ternary system (H₂O → crit)

H₂O ← light compd.

HDO ← 0.015% natural conc.

HTO

Ignore HDO → Binary Sep. (H₂O - HTO)

$$\alpha = \frac{P_1}{P_2}$$

$$\frac{P_2}{P} = \frac{P_{HTO}}{P_{H_2O}} \approx 0.95$$

Vacuum fractionation @ ~55°C

(D.G. Jacobs "Source of Tritium..." p.36)

$$\log \alpha = \log (1/0.95) = 0.0513$$

$$\text{So } n (0.0513) = \log \left[\frac{(x_1/x_2)_p}{(x_1/x_2)_o} \right] = \log \left[\frac{(10/1)}{(1/10)} \right] = \log 100$$

$$n = \frac{\log 100}{0.0513} = 89.8 \rightarrow 90 \text{ ideal stages For 10-fold conc.}$$

May not be "wise" to do anything with "stock UH" stream because of Pu and Fp in solution. could be very "hot" if water evaporated. Also — cannot go to 100% UH because tritium could get trapped in the •6H₂O. Also to drive water out of the UH crystals, requires much energy and compound would decompose (ex. denitrate) before water driven-off.

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essentially the system is closed with recycling of concentrates, the streams of ^3H will build-up until $\text{INPUT} = \text{OUTPUT}$.

ID: not to be recycled - has smallest amount of ^3H in stream.

Do something with AAD $\rightarrow$ crib which will be routed to 1C col. eventually.

If take-off @ 10 gal/min - could concentrate 10-fold in ~90-ideal stages using vacuum distillation ($T \approx 55^\circ\text{C}$)

Rich Pease recommends:

If we vacuum distillate and try electrolysis (which is a good process but expensive) could get as concentrated as want.

Also (in my tech. paper) $\rightarrow$ suggested to do something to product (UNH) to get rid of HTO. (U-plant takes product and converts to UO_2 and sends water to recover acid - but some of water is released)

UNH - PRODUCT

7/29/70

BASIS: irradi. - 900 MWd/TU

get 0.96 mg ^3H /TU

$\therefore 3.6 \text{ CF} \Rightarrow 28.8 \text{ mg } ^3\text{H}/\text{day}$ into plant.

NO. 001% ^3H in $\rightarrow$ UNH prod.

OR 0.288 $\mu\text{g } ^3\text{H}/\text{day}$ in prod

OR 2.01 (10^{-5}) $\mu\text{g } ^3\text{H}/\text{gal}$

= 20 $\mu\text{g } ^3\text{H}/\text{gal}$.

Flow 64 = 14,340 gal/day

Temp 50°C

sp gr 1.68

UNH 2.12 M 1064 g/L

HNO_3 0.06 3.78

Pu ~ 3 ppb NO

Np $\sim 2 \times 10^{-5} \text{ g/L}$ NO

fp 2400 ~ 20 $\mu\text{Ci}/\text{gal}$ NO
Ru ~ 5 NO

water 612.2 g/L $\Rightarrow (33 (10^6)) \text{ g/day}$
OR 8719 gal/day water in product.

J. Akamine

July 31, 1970

PROCESS 24D $\Rightarrow$ vacuum distillation.

$$\alpha = 1.1275 \text{ @ } 12.7^\circ\text{C}$$

$$= \frac{P_{H_2O}}{P_{HTO}}$$

$$\Rightarrow p = 11.013 \text{ mm Hg}$$

{ Van Hook,
"V.P. of isotopic
waters + brs"

relative Volatility = α

$$\alpha = \frac{y_A (1 - x_A)}{x_A (1 - y_A)}$$

where, x_A & y_A are T_A
equilibrium
(Treybal, p. 285)

Enriched water: $A = HTO$

x_A = mole fr. HTO in liq.

y_A = mole fr. HTO in vapor in equil. w/ x_A

$$x_A \sim y_A \sim 0$$

$$\alpha \approx \frac{y_A (1)}{x_A (1)}$$

$\Rightarrow$

$$y_A = \alpha x_A$$

{ straight line for
equil. diagram
plotted

$$\alpha_T = 1/\alpha_H = 0.88692 \text{ @ } 12.7^\circ\text{C}$$

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J. K. Kamine

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8/3/70

(Perry's Ch. E. H. H. p. 13-28)

COLBURN METHOD FOR PARTIAL REFLEX:

Enriching H_2O
Stripping H_2O

WANT: Reflux Ratio $\frac{L_0}{D} \rightarrow \infty$

Method assumes: Equal molar overflow exists
Slope of vapor-liquid equil. relationship $dy/dx (=m)$ is const.

For enriching cal.

$$n \log \frac{1}{x} = \log [(1-x)M + \lambda]$$

where, n = no. of theor. plates

$$\lambda = mV/L$$

M = conc. limits betw. which it is desired to know n

$$M = \frac{(1-y_0) - m(1-x_p)}{(1-y_0) - m(1-x_p)}$$

where, y_0 = vapor conc. @ lower pt. in col.

y_p = " " " upper " " "

x_p = op. line pt. corresponding to y_p

(if upper pt. is chosen as top of col. & if a tot. condenser is used, $y_p = x_p = x_D$)

For tritiated water: (Treybal, R.E. Mass Transfer Operations)

$$\alpha_A = \frac{y_A(1-x_A)}{x_A(1-y_A)}$$

$$\alpha = \text{volatility} = \frac{1}{n} \frac{P_A}{P_B}$$

$A = H_2O$

$B = HTO$

$$\frac{\alpha_A(1-y_A)}{y_A} = \frac{(1-x_A)}{x_A}$$

if $y_A \approx x_A \approx 1$
then,

$$\alpha_A(1-y_A) \approx (1-x_A)$$

$$\alpha_A y_A = \alpha_A - 1 + x_A$$

$$y_A = 1 - \frac{1}{\alpha_A} + \frac{1}{\alpha_A} x_A = 1 - \frac{1}{\alpha_A} \frac{(1-x_A)}{x_B}$$

$$\therefore \frac{dy_A}{dx_A} = \frac{1}{\alpha_A}$$

$$R_D = \frac{L}{D} = \text{reflux ratio}$$

$$V = D + L$$

$$R_D = \frac{L}{V-L}$$

$$\frac{V}{L} - 1 = \frac{1}{R_D}$$

$$\lambda = \frac{mV}{L} = m \left(\frac{R_D + 1}{R_D} \right)$$

amine

For stripping section:

$$n \log \lambda = \log \left(M - \frac{M-1}{\lambda} \right)$$

where $M = \frac{x_p - x_0/m}{x_b - x_0/m}$

and x_0 = liq. conc. @ lower pt. in col. (= x_w)
 x_p = conc. liq. @ upper pt. of col.

For our problem ($HTO - H_2O$)
 consider H_2O enrichment

Start w/ N300 flows $\rightarrow$ want to concentrate down to
 500 gals/day.

IF all Tritium into sys. w/ U-fuel comes out in 24D
 say, max. conc. in fuel $\sim 1.06 \text{ mg } ^3\text{H} / \text{TU}$ (irrad. 1000 MWd/t)

24D: (F) (assumed)
 (300 flows) (3.6 CF) = 61,203 gals/day $\rightarrow$ 46.7 gal/min.

(1.06 mg $^3\text{H} / \text{TU}$) (3.6 CF) = 31.8 mg $^3\text{H} / \text{day}$ $\rightarrow$ $10.6 (10^{-3}) \text{ g-moles } ^3\text{H} / \text{day}$

Bottoms (W):
 (500 gals/day) = 2.232 flows.

Distillate (D):
 $F = D + W \Rightarrow D = 297.768 \text{ flows}$

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$$\bar{z}_F' = \frac{31.8 \text{ mg}^{\text{H}}/\text{day}}{67,203 \text{ gals/day}} = 4.73193 (10^{-7}) \text{ g}^{\text{H}}/\text{gal.}$$

$$\bar{z}_F = \frac{1.57731 (10^{-7})}{210.277} \frac{\text{g-moles HTO}}{\text{g-moles H}_2\text{O TOTAL}} = 7.501083 (10^{-10})$$

$$x_D' = 0.9 \text{ MPC} = 0.9 (3 \times 10^{-3}) \mu\text{Ci/ml} \left(\frac{\text{g HTO}}{1450 \text{ Ci}} \right) \left(\frac{\text{Ci}}{10^6 \mu\text{Ci}} \right) \left(\frac{\text{mole HTO}}{20 \text{ g HTO}} \right)$$

$$= 3.523964 (10^{-9}) \text{ g-moles HTO/gal.}$$

$$\underline{x_D} = \frac{3.523964 (10^{-9})}{210.277} = 1.67586135 (10^{-11})$$

$$\boxed{\bar{z}_F' F = x_D' D + x_W' W}$$

$$\therefore x_W' = \frac{\bar{z}_F' F - x_D' D}{W} = \frac{1.577 (10^{-7}) (300) - 3.523964 (10^{-9}) (297,768)}{3,232}$$

$$= 2.0721768 (10^{-5}) \text{ g-moles HTO/gal}$$

$$\underline{x_W} = 9.854474 (10^{-8})$$

$$\boxed{\text{HTO Recovery} = \frac{W x_W}{F x_F} (100\%)}$$

$$= 97.74 \%$$

Using TOT. REFLEX eqns from Perry's (B-23)

$$n \log \alpha = \log \left[\frac{(x_1/x_2)_P}{(x_1/x_2)_D} \right] = \log \left[\frac{x_W'}{x_D'} \right]$$

$$n = 72.33 \rightarrow \boxed{73 \text{ ideal stages}}$$

IF Feed is sat'd vapor: $y_F = \bar{z}_F \Rightarrow x^* = 1.778 \times 10^{-7}$

$$\text{Then, } \frac{R_{\min}}{R_{\min} + 1} = \frac{\frac{y_F}{x_D} - 0.0357 (10^{-7})}{\frac{1.778 (10^{-7})}{x^*} - 0.0357 (10^{-7})} = 0.8846$$

$$\therefore R_{\min} = 7.668$$

$$y_F = \frac{R_D x_F}{R_D + 1} \quad \frac{x_D}{R_D + 1} \quad \text{(oper. line for enriching section)}$$

J. Akamine

8/4/70

Wrote a computer code to use COBURN PARTIAL
REFLUX EQNS, but it didn't give the
desired results since the input data
are smaller than 10^{-7} and some
calcs involve quantities too small
for the computer to handle correctly.

Scratched!!

ing Colburn eqns (Perney the Hdbk)

For $R = 10$

$\alpha = 1.1275$

Menrich = 94.97 $\rightarrow$ 95 ideal stages
Nstrip — cannot be calc'd.

(THCOL)

8/6/70

ch Please write a computer code to
do a plate-by-plate calculation
for a MULTI-COMPONENT distillation col.
(eqns used from perney 13-31(?))

in THCOL and get some results.

Needs some debugging. When using high L/V
reflux ratios, $X_{w,i}$'s are negative and greater than 1.00.

8/10/70

Worked on THCOL. Chngd name to STILL. Still needs
debugging.

Program still gives negative X_{w2} for $R_o = 10$ and
 $X_{w3} > 1.0$.

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J. Okamene

8/11/70

Reading: W. Jost & Diffusion in Solids, Liquids, Gases, Academic Press,
Inc., N.Y., 1960 (TID 24635, p. 40, ref. 19)
Ch. 12 "Thermal Diffusion" p. 489-532

p. 519 VI Thermal diffusion constants for mixtures of isotopes.

Rigid elastic spheres:

$$\alpha = \frac{105}{118} \frac{m_2 - m_1}{m_2 + m_1}$$

$\{\alpha = \text{some const. of diffusion}\}$

for $m_2 = 20$ (HTO)
 $m_1 = 18$ (H₂O) $\} \alpha = 0.0468332$

for $m_2 = 4$ (HT)
 $m_1 = 2$ (H₂) $\} \alpha = 0.296610$

Reading: D.G. Jacobs "Sources of Tritium and Its Behavior upon Release
to the Environment" ORNL, Tenn. (TID-24635 (203))
1968.

p. 35 Distillation.

p. 36 (fig. 4.1) plotted data on relative vapor press. of HTO-H₂O
As temp. incr. — sys. decr. @ 100°C $\alpha = 1.036$
2 rel. volatility

Recommends vacuum distillation.
Zel'vinskii et al.³² → opt. fractionation of tritium w/ respect
to water (for identical times of distillation) achieved
@ 100 — 120 mmHg. (water Bp. @ 52 — 55°C)

8/12/70

Had problems with teletype and computer. Revised
and modified program STILL — negative numbers still
appear for $R=10$.

J. Ekamine

8/13/70

plotted Reflux Ratio vs. No. of theoretical stages
to produce 0.9 MPC in distillate.
Error in plot considered since computer
code is in error but plot gives
us some kind of "feel" for systems
considered.

~~XXXXXXXXXX~~

Input stream is 75 — 300 flows.

Distillate is 0.9 MPC.

Store is ~ 500 gals/day

(x 200 opn. days → 100,000 gals/yr)

Using info. from SECRET document : USAEC R&D Report (May 1968)
BNWL-784 p. 71-2

	MND/t	Tritium content / t		INPUT / day	
		mg	g	mg T	H ₂ O (gmols)
3CF (14.99 T/day)	640	0.68	6.6 (164.9 g/day)	16.99	(5.663 (10 ³))
	750	0.80	7.8 (194.9)	19.99	(6.66)
	900	0.96	9.3 (232.4)	23.99	(8.00)
2.5 C.F. Clad (20,825)	900	0.96	9.3 (193.7)	19.99	(6.66)
	1000	1.06	10.4 (216.6)	22.07	(7.36)
1 C.F. (8.33) 3 IRC Clad	1200	1.28	12.4 (103.3)	10.66	(3.55)
	2200	2.33	22.6 (188.3)	19.41	(6.47)
	2200	2.33	22.6 (")	"	(")
	2100	2.22	21.5 (179.1)	18.49	(6.16)

300 flows = 56603 gals/day (3CF)
46669 (2.5)
18668 (1)

∴ Assume with condensate recycling — INPUT (fuel) = OUTPUT (300 fl)

$$F = D + W$$

$$z_F F = x_D D + x_W W \Rightarrow x_W = \frac{z_F F - x_D D}{W}$$

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$$= CF (18667.53 \text{ gal/day})$$

$$900 \text{ Flows} = (900)(CF)(13084.5) \text{ g-moles H}_2\text{O} = 3.925 (10^6)(CF) \text{ g-moles H}_2\text{O}$$

MAN/E	HTO	$Z_F (10^9)$	$W (500 \text{ gal/day})$	$X_W (10^8)$	$X_W (10^8)$	% Recovery
640	✓	0.48096	2.678	5.33402	5.36930	99.9%
750	3CF	0.56561	2.678	6.27282	6.31758	
900		0.67941	2.678	7.53491	7.59241	
900	2.5	0.67873	3.214	6.272027	6.31990	
1000	*	0.75006	3.214	6.931170	6.98571	
1200	✓	0.90446	8.035	3.343181	3.37086	
2200	1.0	1.64841	8.035	6.09306	6.14852	
2100	*	1.56943	8.035	5.80113	5.85364	

$$D = F - W = 300 - W$$

$$Z_F = \frac{\text{g-moles HTO}}{\text{g-moles tot. water}} = \frac{\text{g-moles HTO}}{\text{g-moles H}_2\text{O}} \quad \text{g-moles H}_2\text{O} = (\text{Flows})(CF)(62.225 \frac{\text{gal}}{\text{day}})(210.28)$$

$$\rightarrow X_D = 0.9 \text{ MPC} = 2.7 \times 10^{-3} \mu\text{Ci/cc} = 3.523965 (10^{-9}) \frac{\text{g-moles HTO}}{\text{gal H}_2\text{O}} = 1.67586 (10^{-11}) \frac{\text{g-moles HTO}}{\text{g-mole H}_2\text{O}}$$

$$\% \text{ Recovery} = \frac{\text{RECOVERED}}{\text{IN}} (100\%) = \frac{WX_W}{FZ_F}$$

$$\text{if } 99\% \text{ recovery wanted: } X_W = (0.99)(FZ_F)/W$$

$$75 \text{ FLOWS} = (CF)(4666.9 \text{ gal/day}) = (CF)(9.814 (10^5)) \text{ g-moles H}_2\text{O}$$

MAN/E	$(10^8) Z_F$	99% Rec	W	XW	50 gal/day	100 gal/day
640	0.192344	142815			ICF = 0.8035 flows	1.6070
750 (3CF)	0.22621	1.67961			2.5 = 0.3214	0.6428
900	0.27172	2.01752			3 = 0.2678	0.5356
900 (2.5)	0.27145	2.01552				
1000	0.29998	2.22735				
1200	0.36173	2.68585				
2200 (1.0)	0.65926	4.89501				
2100	0.62767	4.66045

IF: Ignore HTO leaving in other streams and assume all HTO in goes out in 75 flows.

J. Alkamine

8/18/70

$$\rho_{H_2O} = 8.337 \text{ lb/gal.}$$

$$\text{storing condensate @ } \$1.00/\text{gal.} \Rightarrow (500 \text{ gal/day}) (\$1.00/\text{gal}) = \$500/\text{day}$$

$$\text{Reflux Ratio} = 20 = \frac{L}{D} \rightarrow V = 21 \quad (V = L + D)$$

$$\text{steam cost } \$1.00/1000 \text{ lb}$$

$$(1 \text{ gal } H_2O) (8.337 \text{ lb/gal}) (21 \frac{\text{lb steam}}{\text{lb liq}}) (1.333) = 232.9 \text{ lb steam}$$

$$\$0.24/\text{gal. @ } R_p = 20$$

$$\$13,460/\text{day @ 300 flows (3CF)}$$

$$R_p = 30$$

$$V = 31$$

$$(1 \text{ gal}) (8.337) (31) (1.33) = 344$$

$$\Rightarrow \$0.35/\text{gal @ } R_p = 30$$

$$\$19,600/\text{day}$$

$$R_p = 40$$

$$V = 41$$

$$\text{Cost} = \frac{454.6 \text{ lb}}{1000}$$

$$= \$0.46/\text{gal @ } R_p = 40$$

$$\$25,800/\text{day}$$

Condenser duty:

$$R = 20$$

$$V = 21$$

$$V H_v = Q_c + L H_L + D H_D = Q_c + H_L (L + D)$$

$$\frac{L}{D} = 20$$

$$V = L + D$$

$$L = 20D$$

$$V = 21D$$

$$V H_v = Q_c + H_L V$$

$$Q_c = \underbrace{V(H_v - H_L)}_{\text{latent}}$$

$$@ T_c = 27^\circ\text{C}$$

From Perry's (3-114) for water

$$\lambda_v = 1020 \text{ Btu/lb} = 580 \text{ cal/gm.}$$

For 1 gal. H_2O (vapor)

$$Q_c = 8503.74 \text{ Btu} = 2.195 (10^6) \text{ cal.}$$

For 300 flows in — 297.3 = D

$$Q_c \approx 1.37 (10^{10}) \text{ cal/day}$$

$$\approx 5.31 (10^7) \text{ Btu/day}$$

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8/13/70

$$\left. \begin{array}{l} F=300 \\ D=297.3 \\ W=2.7 \end{array} \right\} 3CF$$

$$\frac{D}{W} = 110$$

$\Rightarrow$ store 1 gal of W @ 7/100
Expend \$27.53 for steam w/R=20

8/14/70

Worked on STILL.

Met a man - Bob Keil (can never remember names) who suggest blowing up the stack (what we've been avoiding to even think about)

8/18/70

Think I have STILL "debugged" - nothing wrong with the computer code, just the magnitude of one of the inputs - Y. (the fraction of H₂O allowed to go overhead).

Ran 3 cases:

$\left. \begin{array}{l} \text{ratios} \\ 2.30, 40.50 \\ 0.22 \end{array} \right\}$	1) Al clad:	1000 MWd/t	
	F=300	W=3.214	$Z_F(H_2O) = 7.5006 E-10$
	2) Al clad:	1000 MWd/t	
	F=75	W=3.214	$Z_F(H_2O) = 7.5006 E-10$
	3) Zirc clad:	2100 MWd/t	
	F=300	W=8.075	$Z_F(H_2O) = 1.56943 E-9$

Tried running Zirc clad F=75 but not sure it works - got total tower of 1 stage. (Discussed this in input)

Plotted reflux ratio vs. Feed plate and Total no. of theoretical stages for the 3 cases.

J. Akamine

8/20/70

N/200 flows of process Condensates (TOTAL)
(all @ $T = 50^{\circ}\text{C} = 122^{\circ}\text{F}$)

OF $h_{fg} \approx 1113.7 - 87.92 = 1025.78 \text{ Btu/lbm}$ {Purveys}

1200 flows $\approx 22 (10^6) \text{ gal/day} \approx 200 (10^6) \text{ lb/day}$

To vaporize: need $\approx 206 (10^9) \text{ Btu/day}$
 $206 (10^9) \text{ Btu/day} \approx 60 (10^9) \text{ kWh/day} \rightarrow \textcircled{\$600,000}$
@ 1¢/kWh (electrical)

OR $\# \text{ steam} / \# \text{ water}$ @ $\$100 / 1000 \# \text{ steam}$

1200 flows $\approx 200 (10^6) \text{ lbs/day}$ (1.33 safety factor)

$\approx 266 (10^6) \text{ lb steam/day}$
 $266 (10^6) \# \left(\frac{\$100}{1000 \#} \right) = \$266,000 \rightarrow \textcircled{\$300,000}$
(thermal)

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8/24/70

Total process condensates:

AAD	57.7	flows
14D	350	
24D	317	
PAW	5	
BWD	290 (?)	
Chem Sewer	141.8	

1161.5 $\rightarrow$ 1200 flows

Steam repts:

$$(98\% \text{ } ^3\text{H in condensates})(1.06 \text{ mg } ^3\text{H/TU @ 2.5 CP})$$

$$= 21.63 \text{ mg } ^3\text{H} = 2.23 \text{ Ci/day}$$

$$1200 \text{ flows} = 1.87 (10^5) \text{ gals/day} = 7.07 (10^5) \text{ L/day}$$

$$\therefore \text{concentration of process condensates combined}$$

$$= 3.15 (10^{-5}) \text{ Ci/L} = 3.15 (10^{-2}) \mu\text{Ci/cc.}$$

$$\text{MPC for air} = 2 \times 10^{-7} \mu\text{Ci/cc}$$

Let X = water of dilution needed to obtain MPC

$$\frac{(2.23 \text{ Ci/day})(10^6 \mu\text{Ci/Ci})}{(7.07 (10^8) \text{ L/day of condensates} + X \text{ cc/day})} = \text{MPC} = 2 \times 10^{-7} \mu\text{Ci/cc}$$

$$X = \frac{2.23 (10^6)}{2 (10^{-7})} - 7.07 (10^8) \approx 1.11 (10^{13}) \text{ cc/day}$$

$$\approx 2.9 (10^9) \text{ gal/day} \approx \underline{1.9 (10^6) \text{ flows dilution water}}$$

J. Alkamine

8/25/70

$$68.317 \text{ kcal/g-mole} = -\Delta H_c \text{ for } \text{H}_2$$

$$F = 300 \text{ flows} \rightarrow W = 3.2 \text{ flows}$$

ELECTROLYZED (decomposed)

$$\Delta H_c \text{ needed} = (300 - 3.2)(\Delta H_c) = 6.63 (10^8) \text{ kcal/day}$$

$$= 3.2 (10^4) \text{ KW} = \underline{32 \text{ MW}}$$

Jean Alkamine

E/26/70

looked at H_2S-H_2O exchange processes

Ref. 1) Bibbington, W.P. & J.E. Proctor, W.C. Scotter & V.R. Thayer
 "Production of heavy water in the United States of America"
 3rd Int'l Conf. on the Peaceful Uses of Atomic Energy, Vol. 12
 p. 334 - 339

2) Rae, H.K. "Chemical Exchange Processes for Heavy Water"
 AECL-2555 (Conf. 660516-2) May 1960 (Chalk River,
 Ontario)

3) Jackson, D.P. "Equilibrium Constants for GS
 process reactions involving Tritium
 compounds" AECL-3322 Vol 3-69
 (Chalk River, Ontario) microfiche

$$\alpha = \text{separation factor} = K \left(\frac{m}{n} \right)$$

from Jackson

$$K = \frac{P[H_2S]/P[HTS]}{P[H_2O]/P[HTO]}$$

	@ T°C	K	(1/K = 1/K_T = m_T)
pt. of d. for DO-H ₂ S	30	2.777	0.306
	130	2.146	0.466

for (A) $m=n=2$ so $\alpha = K$

Then, $Y = \frac{1}{\alpha_T} X = m_T X$

OR, $\frac{P[HTS]}{P[H_2S]} = \frac{1}{K} \frac{P[HTO]}{P[H_2O]} = m_T \frac{P[HTO]}{P[H_2O]}$

plotted $Y = m_T X$ on graph paper.

P_T (opt.) = 300 psi

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Isotopic exchange reactions using H_2O-H_2S is very similar to gas absorption, thus drawing equilibrium lines and operating lines, we can "step-off stages" for the reaction tower.

H_2S slightly soluble in H_2O ~ 0.012 mole % H_2S in H_2O
so ignored solubility effect.

$$3.2 \text{ flows} \approx 500 \text{ gpd} = 0.346 \text{ gpm}$$

$$300 \text{ flows} \approx 47,000 \text{ gpd} = 32.41 \text{ gpm} = 15.01 \text{ lb-moles/min}$$

$$\frac{L}{G} \approx 0.416$$

$$L = 15.01 \text{ lb-moles/min} \Rightarrow G = 36.08 \text{ lb-moles/min} \\ = 622 \text{ cfm}$$

$$500 \text{ gpd} \approx 420 \text{ T/yr} @ 200 \text{ op. days / yr.}$$

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8/26/70

Questions posed:

1) What does the equipment look like for various processes?

a) distillation

b) electrolytic

c) $H_2S - H_2O$

d) etc.

2) Sketch them as to size, etc.

H_2S exchange very promising!

J. Akamine

End of work performed by
J. Akamine

J. Akamine
9/10/70

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DATE

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4 / 20 / 94

END

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