Numerical study on extraction of tritium generated in HMR by way of system composed of EXEL-process and thermal diffusion column cascade

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Abstract A new tritium extraction system composed of a trickle-bed hydrogen/water isotopic exchange column using a hydrophobic Pt-catalyst combined with an SPE-water electrolyser (EXEL-process) and a thermal diffusion column cascade was proposed for the removal of tritium from heavy water irradiated in HMR ((Heavy Water Moderated Power Reactor), volume of heavy water = 140 m^3 and mean neutron flux = 5×10^{13} n/cm^2s). Numerical study on the extraction of tritium from the heavy water was carried out and the dimensions of proposed system were determined under the conditions that the concentration of tritium in the heavy water was kept less than 2.5 Ci/l_{HW}. The calculation results indicated that the proposed system was designed practically.

Key words chemical exchange reaction • heavy water • isotope separation • thermal diffusion • trickel-bed reactor • tritium

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Introduction

Tritium is produced in the heavy water of HMR (Heavy Water Moderated Power Reactor) principally by the following nuclear reaction, D(n,γ)T. If the tritium in the heavy water is not removed, the concentration of tritium reaches a saturated value (30 to 40 Ci/l_{HW}), which depends on the neutron flux intensity. Such high concentration of tritium is dangerous for reactor operators, because of increasing their irradiated dose. So tritium has to be removed from the heavy water.

The tritium extraction system (TES) for the removal of tritium from heavy water, which is operated at present in France and Canada, is composed of two isotopic separation processes:

– chemical exchange process using Pt-loaded silica gel as a catalyst at 473 K,
– cryogenic distillation process of tritiated deuterium [1, 2, 4].

In the first process, tritium is separated according to an isotopic exchange reaction, $DTO(g)+D_2(g) = D_2O(g)+DT(g)$ (K = 0.82 at 473 K). Then, the process has to be operated at high temperature. On the other hand, the second process is operated at very low temperature, 25 K [1]. The present system is too uneconomical and should be improved reasonably from the viewpoint of energy saving. Thus, the authors propose a new TES, which is composed of

– chemical exchange process using 0.5wt%Pt/SDBC as a catalyst at 353 K (SDBC: styrene-divinylbenzene copolymer, 4 mm in diameter),
– thermal diffusion process of tritiated deuterium gas.

In the first process, an isotopic exchange reaction, $D_2O(l) + DT(g) = DTO(l) + D_2(g)$, proceeds at 353 K, which is lower than that of the conventional process. This reaction is carried out in the trickle-bed isotopic exchange column with the pressurized SPE-type water electrolyser (EXEL-process, the pressure of electrolyser = 5 to 40 atm). The thermal diffusion (TD) process is a column cascade consisted of several thermal diffusion columns and a part of tritiated deuterium gas is transferred from the electrolyser to the TD process [3, 7]. The schematic diagram of proposed TES is shown in Fig. 1.

In order to maintain the tritium concentration of heavy water to less than the safe level (2.5 Ci/l_{HW}), the removal of tritium by the TES is started when the concentration of tritium exceeds 2.5 Ci/l_{HW}. As shown in Ref. [4], the generation of tritium in the heavy water of HMR is represented as

$$(1\text{-}1) \quad \left((d/dt)(VA)\right)_{t=t_c} = V \cdot A_s \cdot \lambda \exp(-\lambda t_c)$$

where t_c denotes the operation time of HMR until the concentration of tritium in heavy water reaches 2.5 Ci/l_{HW}. Therefore, total mass balance of proposed TES is represented as

$$(1\text{-}2) \quad F \cdot A_c - (F - P) \cdot (A_c / q_s) = V \cdot A_s \cdot \lambda \exp(-\lambda t_c)$$

where F, A_c and q_s denote the draw-off rate of tritiated heavy water from HMR to the isotopic exchange column, the concentration of tritium maintained by the TES (2.5 Ci/l_{HW}) and the tritium depletion factor, respectively. Assuming $(1 - (P/F)) \sim 1$ and using $A_c = A_s (1 - \exp(-\lambda t_c))$, we get

$$(1\text{-}3) \quad F = \lambda V \left((A_s - A_c)/A_c\right)\left(q_s/(q_s - 1)\right)$$

Since A_s ($= \gamma \cdot \phi \cdot 10^{-12}$), λ and V are given as 25 (Ci/l_{HW}), 6.272×10^{-6} (h^{-1}) and 140×10^3 (l_{HW}), respectively, Eq. (1-3) is expressed by

$$(1\text{-}4) \quad F = (0.892)\left(1 - (A_c/A_s)\right)/(A_c/A_s)\left(q_s/(q_s - 1)\right) \quad [l_{HW}/h]$$

Assuming that $A_c = (1/10) A_s$ and $q_s = 10$, the following values, $F = N_F = 8.92\ l_{HW}/h = 0.4956$ (kmol /h), $X = A_c = 2.5$ $Ci/l_{HW} = 7.5\times10^{-7}$ (T – atom fraction), $R = N_F - P_T = 0.4956$ kmol/h, $X_R = (1/10)\ X = 7.5\times10^{-8}$ (T – atom fraction), are obtained. Assuming that the tritium atom fraction X_P is given as 0.1 from the following relation, $FX = R \cdot X_R + P_T \cdot X_P$, we get $P_T = 3.3453\times10^{-6}$ kmol /h.

In this paper, the numerical calculation to make a flow diagram of proposed TES was carried out and the dimensions of each process in the TES were evaluated. From these results, the applicability of proposed TES to the removal of tritium from heavy water irradiated in HMR was discussed.

Theory

Isotopic exchange of D/T between $D_2O(l)$ and $D_2(g)$ in trickle bed column

The mass balance of tritium in the trickle bed reactor is expressed as follows

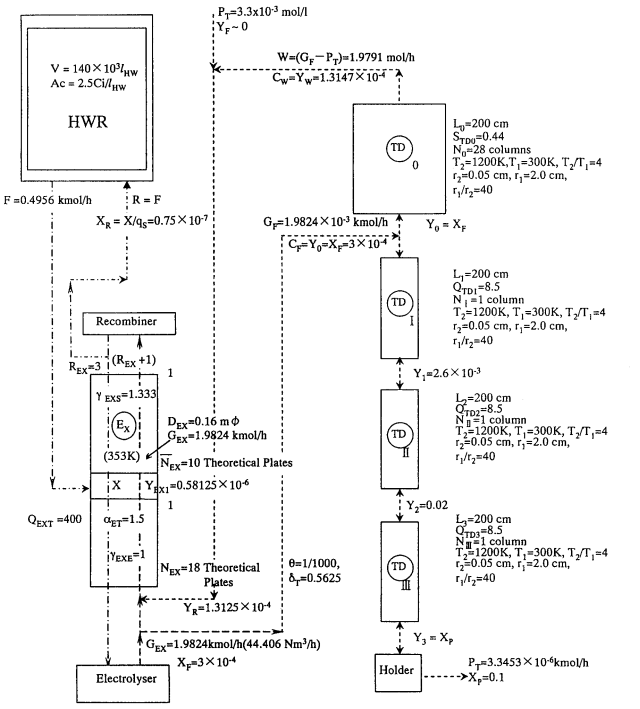

Fig. 1. Schematic diagram of tritium extraction system composed of D/T-exchange reaction column and thermal diffusion column cascade.

$$(2\text{-}1) \quad \frac{G_F}{F} = \left(\frac{X}{X_F}\right) \cdot \frac{\left(1 + \frac{1}{q_S}\right)}{\delta_T} = \left(\frac{1}{Q_{EXT}}\right) \cdot \frac{\left(1 + \frac{1}{q_S}\right)}{\delta_T}$$

where Q_{EXT} denotes the enrichment factor of tritium. The number of theoretical plates of the stripping section of EX column (see Fig. 1), $\overline{N}_{EX}$ is calculated by the following expression [8]

$$(2\text{-}2) \quad \overline{N}_{EX} = \frac{\ln\left\{\frac{\left(X - \frac{X}{q_S}\right)\left(\frac{\alpha_{ET}}{\gamma_{EXS}} - 1\right)}{(\alpha_{ET} - 1)\left(\frac{X}{q_S}\right)} + 1\right\}}{\ln\left(\frac{\alpha_{ET}}{\gamma_{EXS}}\right)}$$

where $\gamma_{EXS} = (R_{EX}+1)/R_{EX}$.
Then, the reflux ratio of EX column, R_{EX}, is calculated by the following expression,

$$\frac{X\left(1 - \frac{1}{q_S}\right)\left[\frac{\alpha_{ET} \cdot R_{EX}}{(R_{EX}+1)} - 1\right]}{(\alpha_{ET} - 1)\left(\frac{X}{q_S}\right)} + 1 > 0$$

Therefore,

$$(2\text{-}3) \quad R_{EX} > \frac{\left(1 - \frac{\alpha_{ET}}{q_S}\right)}{(\alpha_{ET} - 1)}$$

From the definition of R_{EX}, the generation of deuterium gas at the electrolyser is described as

(2-4) $$G_{EX} = F\left(R_{EX}+1\right)$$

Accordingly, the feed rate of deuterium to the thermal diffusion cascade, G_F, is described as

(2-5) $$G_F = \theta G_{EX} = \theta \cdot F\left(R_{EX}+1\right)$$

where θ denotes the cut. By substituting Eq. (2-5) into Eq. (2-1), the cut is expressed as

(2-6) $$\theta = \left(\frac{1}{Q_{EXT}}\right)\cdot\frac{\left(1-\frac{1}{q_S}\right)}{\delta_T\left(R_{EX}+1\right)}$$

The number of theoretical plates in enriching section, N_{EX}, can be calculated as [8]

(2-7) $$N_{EX} = \frac{\ln\left\{\frac{X\left(Q_{EXT}-1\right)\left(\alpha_{ET}-1\right)}{\alpha_{ET}\cdot Y_{EX1}-X}+1\right\}}{\ln\left(\alpha_{ET}\right)}$$

where

$$Y_{EX1} = X\left\{1-\frac{F}{G_{EX}}\left(1-\frac{1}{q_S}\right)\right\} = X\left\{1-\frac{1}{\left(R_{EX}+1\right)}\left(1-\frac{1}{q_S}\right)\right\}$$

Thermal diffusion column

Thermal diffusion column constants, H, K_c, K_d and K_p, are expressed as follows [10],

(2-8) $$H = \left(\frac{2\pi}{6!}\right)\left(\frac{\alpha_T\cdot\rho^2\cdot g}{\eta}\right)_{T1} r_1^4\, h\left(\frac{T_2}{T_1},\frac{r_1}{r_2}\right) \quad [g/s]$$

(2-9) $$K_c = \left(\frac{2\pi}{9!}\right)\left(\frac{\rho^3 g^2}{\eta^2 D_{T/D}}\right)_{T1} r_1^8\, k_c\left(\frac{T_2}{T_1},\frac{r_1}{r_2}\right) \quad [g\ cm/s]$$

(2-10) $$K_d = 2\pi\left(\rho\, D_{T/D}\right)_{T1} r_1^2\, k_d\left(\frac{T_2}{T_1},\frac{r_1}{r_2}\right) \quad [g\ cm/s]$$

(2-11) $$K_p = (0.3)K_c$$

where α_T denotes the thermal diffusion coefficient and is given as 0.042 [10]. The diffusion coefficient of DT in D_2 gas is described as [5]

(2-12) $$\left(D_{T/D}\right)_{\substack{300^\circ K\\ 1\ atm}} = \frac{0.00070}{\left[\left(\frac{T_c}{P_c}\right)_{TD}^{1/3}+\left(\frac{T_c}{P_c}\right)_{D2}^{1/3}\right]}\cdot\sqrt{\frac{1}{M_{TD}}+\frac{1}{M_{D2}}} = 0.8886 \quad [cm^2/s]$$

When the deuterium gas is drawn off, the tritium transport equation for the 0-th column is given as

(2-13) $$\left(C_W/C_F\right) = \frac{\left(1-\gamma_0\right)}{q_{eo}^{(1-\gamma_0)}-\gamma_0}$$

where $C_W = Y_W$, $C_F = X_F$, $\sigma_W = (G_F - P_T)$ (1.1111) [g/s], $\gamma_0 = (\sigma_W/N_0H_0)$, $q_{eo} = \exp\left(\frac{L_0H_0}{K_0}\right)$

The tritium transport equation for the first column is given as follows,

(2-14) $$\left(\frac{C_{1B}}{C_F}\right) = \frac{\left(1+\gamma_1\right)q_{e1}^{(1+\gamma_1)}}{1+\gamma_1\cdot q_{e1}^{(1+\gamma_1)}}$$

where $\gamma_1 = \frac{\sigma_P}{N_1\cdot H_1}$, $q_{e1} = \exp\left(\frac{L_1H_1}{K_1}\right)$, $\sigma_P = P_T$.

The tritium transport equation for the the n-th column is described similarly as

(2-15) $$\frac{C_{nB}}{C_{(n-1)B}} = \frac{\left(1+\gamma_n\right)q_{en}^{(1+\gamma_n)}}{1+\gamma_n\cdot q_{en}^{(1+\gamma_n)}}$$

where $\gamma_n = \frac{\sigma_P}{N_n\cdot H_n}$, $q_{en} = \exp\left(\frac{L_n\cdot H_n}{K_n}\right)$.

From these results, the number of the stages of the thermal diffusion cascade, N_{TDC}, is calculated as

(2-16) $$N_{TDC} = \frac{\log\left(\frac{X_P}{X_F}\right)}{\log\left(\frac{C_{nB}}{C_{(n-1)B}}\right)}.$$

Results of numerical study

Assumptions of calculation

The calculation of mass balance of proposed TES was carried out on the basis of the following assumptions, A_s = 25 Ci/l_{HW} = 7.5×10^{-6} [T – atom fraction], q_s = 10, A_c/A_s = 1/10, $X = (7.5\times10^{-6})(1/10) = 0.75\times10^{-6}$ [T – atom fraction], $X_R = X/q_s = 0.75\times10^{-7}$ [T – atom fraction], Q_{EXT} = 400, θ = 1/1000, temperature of EX column = 353 K (α_{ET} = 1.50), S_{TDO} = 0.44.

Calculation results

The calculation results obtained were summarized as follows,

Eq. (1-4) F = 8.92 l/h = 0.4956 kmol/h,
$R = F$ = 0.4956 kmol/h,

Eq. (2-3) R_{EX} = 3,

Eq. (2-2) $\overline{N}_{EX}$ = 10 theoretical plates,

Eq. (2-7) N_{EX} = 18 theoretical plates,
$Y_{EX1} = 0.58125\times10^{-6}$,
$X_F = X\cdot Q_{EXT} = 3\times10^{-4}$,

Eq. (2-5) $G_F = 1.9824\times10^{-3}$ kmol/h,
$Y_W = S_{TDO}\cdot X_F = 1.3147\times10^{-4}$,
$Y_R = Y_W\cdot W/(W+P_T) = 1.3125\times10^{-4}$,

Ref. [6] P_{opt} = 423 mmHg; h = 0.1282; k_c = 0.01556; k_d = 0.7037; Force Index n = 0.75 (r_1/r_2) = 40; (T_1/T_2) = 4; $H_0 = 4.8530\times10^{-5}$ g/s/column for one column; $K_c = (5.1690\times10^{-3})(2^8)(0.01556)(423/760)^4 = 1.9759\times10^{-3}$ g cm/s/column; $K_d = (9.1386\times10^{-4})(2^2)(0.7037) = 2.5723\times10^{-3}$ g cm/s/column,

Eqs. (2-13), (2-14), (2-15) $q_{eo} = q_{e1} = q_{e2} = q_{e3}$ = 33.459;

Eq. (2-13) γ_0 = 1.66; (C_w/C_F) = 0.44, where $C_w = 1.31\times10^{-4}$ and $C_F = 3\times10^{-4}$ are assumed.

$$N_0 = \frac{(1.9791\times10^{-3})(1.1111)}{(1.66)(4.850\times10^{-5})} = 28 \text{ columns, where}$$

$N_0 = \sigma_W/\gamma_0 H_0$ and
$\sigma_W = (G_F - P_T)$, $C_{1B} = (8.5)(3\times10^{-4}) = 2.55\times10^{-3}$ from Eq. (2-14), where r_1 = 0.1 and q_{e1} = 33.459,

Eq. (2-16) $N_{TDC} = \log(0.1/3\times10^{-4})/\log(8.5) = 3$ stages,

Eq. (2-14) $N_1 = \frac{(3.3453\times10^{-6})(1.1111)}{(0.1)(4.8530\times10^{-5})} = 0.77$ column ~1 column, $C_{2B} = 2.55\times10^{-3}(8.5) = 0.0217$ from Eq. (2-15), where γ_n = 0.1 is assumed and q_{e2} = 33.459, N_2 = 1 column; $C_{3B} = (0.0217)(8.5) = 0.18$ regarded as C_{3B} = 0.1.

If the value of Q_{EXT} is increased, the number of the theoretical plates of the enriching section becomes greater, however, the number of thermal diffusion columns at the 0th-stage becomes smaller. The optimum value of Q_{EXT} can be determined from the economic conditions. The smaller is the cut, the less is the number of the thermal diffusion columns at the 0th-stage. So the cut of 1/1000 is regarded as mechanically appropriate. These calculated values were also shown in Fig. 1.

Conclusions

Concerning the extraction of tritium generated in the HMR (140 m^3 heavy water and 5×10^{13} n/cm^2s), the TES composed of the EXEL-process and the thermal diffusion column cascade (TDC) was proposed and the numerical study on the extraction of tritium from heavy water irradiated in the HMR. The dimensions of proposed system were determined by optimizing the cut (θ) from the EXEL-process to the TDC. The calculation results indicated that the proposed system was designed practically. The dimensions and operating conditions of proposed TES were summarized as follows:

1. The combination of the EXEL-process and the TDC composed of 28 columns for the first stage of stripping (TD_0) and 3 columns for the 3 stages of enriching (1 column for each stage).
2. The details of the thermal diffusion column are as follows: – length of column: 200 cm; – inner radius of column: 20 mm; – outer radius of hot wire: 0.5 mm; – temperature of hot wire: 1200 K; – temperature of cold wall: 300 K.
3. The details of the EXEL-process are as follows: – stripping section: 10 theoretical plates; – enriching section: 18 theoretical plates; – diameter of exchange column: 0.126 m; – length of column = (10+18)(HETP) = 8.4 m, where HETP = 0.3 m [9]; – SPE-water electrolyser; – recombiner.
4. The cut from the EXEL-process to the TDC: 1/1.000.
5. Extraction of tritium: 150.000 Ci/300 d.

Symbols

A: [Ci/l_{HW}] Concentration of tritium in heavy water of HMR at time t $(= A_s(1 - e^{-\lambda t}))$;
A_c: [Ci/l_{HW}] Controlled concentration of tritium in heavy water of HMR $(= (1/10)A_s)$;
A_s: [Ci/l_{HW}] Saturated concentration of tritium in heavy water of HMR;
C: [T – atom fraction] Concentration of tritium;
$D_{T/D}$: [cm^2/s] Diffusion coefficient of DT in D_2 gas (= 0.8886 cm^2/s at 300 K and 1 atom);
F: [kmol/h] Draw-off rate of tritiated heavy water (= 0.4956 kmol/h);
g: [cm/s^2] acceleration of gravity (= 980 cm/s^2);
G_{EX}: [kmol/h] Generation of deuterium from electrolyser;
G_F: [kmol/h] Flow rate of tritiated deuterium gas to TDC-cascade;
H: [g/s/column] Mass transfer coefficient by thermal diffusion and convection;
h: [-] Shape factor concerning H;
K: [g cm/s/column] = $K_c + K_d + K_p$;
K_c: [g cm/s/column] Mass transfer coefficient for counter flow caused by convection;
k_c: [-] Shape factor concerning K_c;
K_d: [g cm/s/column] Mass transfer coefficient for counter flow caused by diffusion;
k_d: [-] Shape factor concerning K_d;
K_p: [g cm/s/column] Mass transfer coefficient for counter flow caused by parasite effect;
L: [cm] Length of thermal diffusion column;
l_{HW}: [l] Liter of heavy water;
M: [g/mol] Molecular weight;
N_{EX}: [-] Number of theoretical plates of enriching section of EX-column;
$\overline{N}_{EX}$: [-] Number of theoretical plates of stripping section of EX-column;
N_{TDC}: [-] Number of stages in thermal diffusion cascade;
n: [-] Stage number of thermal diffusion cascade;
$[P_c]_{TD}$: [atm] Critical pressure of DT;
$[P_c]_{D2}$: [atm] Critical pressure of D_2;
P_T: [kmol/h] Production rate;
Q_{EXT}: [-] T – enrichment factor in EX-column;
Q_{TD1}: [-] T – enrichment factor in TD_I column $\left(= \frac{Y_1}{1-Y_1} \Big/ \frac{X_F}{1-X_F} \approx Y_1/X_F\right)$;
Q_{TD2}: [-] T – enrichment factor in TD_{II} column $\left(= \frac{Y_2}{1-Y_2} \Big/ \frac{Y_1}{1-Y_1} \approx Y_2/Y_1\right)$;
Q_{TD3}: [-] T – enrichment factor in TD_{III} column $\left(= \frac{Y_P}{1-X_P} \Big/ \frac{Y_2}{1-Y_2} \approx \frac{X_P}{1-X_P} \Big/ Y_2\right)$
q_s: [-] Depletion factor of tritium;

q_{en}: [-] Equilibrium separation factor in thermal diffusion column at the n-th stage of TD cascade ($= \exp(L_n H_n/K_n)$);
r_1: [cm] Inner diameter of thermal diffusion column;
r_2: [cm] Diameter of hot wire;
S_{TD0}: [-] T – depletion factor of TD_0 column ($= Y_W/X_F$);
$[T_c]_{TD}$: [K] Critical temperature of DT;
$[T_c]_{D2}$: [K] Critical temperature of D_2;
V: [l_{HW}] Quantity of heavy water filled in HMR ($= 140\times10^3\ l_{HW}$);
W: [kmol/h] Flow rate of T – depleted waste;
X: [T – atom fraction] Concentration of tritium in heavy water;
X_F: [T – atom fraction] Concentration of tritium in feed;
X_P: [T – atom fraction] Concentration of tritium in T – enriched product;
X_R: [T – atom fraction] Concentration of tritium in returned heavy water ($= X/q_s$);
Y_0: [T – atom fraction] Concentration of tritium in deuterium gas fed to TD process;
Y_1: [T – atom fraction] Concentration of tritium between TD_I and TD_{II};
Y_2: [T – atom fraction] Concentration of tritium between TD_{II} and TD_{III};
Y_3: [T – atom fraction] Concentration tritium at the bottom of TD_{III};
Y_{EX1}: [T – atom fraction] Concentration of tritium in deuterium gas at the feed point of EX column;
Y_F: [T – atom fraction] Concentration of tritium in deuterium gas fed to EXEL-process;
Y_W: [T – atom fraction] Concentration of tritium in T – depleted waste from TD_0 column;
α_{ET}: [-] D/T – separation factor $DTO + D_2 = D_2O + DT$;

$$\alpha_{ET} = \frac{[DTO][D_2]}{[D_2O][DT]} \approx \frac{[DTO]}{[DT]};$$

α_T: [-] Thermal diffusion coefficient (= 0.042) [10];
γ: [-] Neutron irradiation factor of deuterium (= 0.5);
γ_{EXE}: [-] Gas/liquid flow ratio in enriching section of EX-column;
γ_{EXS}: [-] Gas/liquid flow ratio in stripping section of EX-column;
γ_n: [-] Parameter of TD column in case of draw-off operation;
δ_T: [-] Value defined as $(X_F - Y_R)/X_F$;
η: [g/cm s] Viscosity of D_2 gas;
θ: [-] Cut;
λ: [h^{-1}] Degradation constant of tritium ($= 6.372\times10^{-6}$);
ρ: [g/cm^3] Density of D_2 gas;
σ_P: [g/s] Draw-off rate in the n-th stage of TD cascade ($= P_T\times1.1111$);
σ_W: [g/s] Draw-off rate in the 0-th stage of TD cascade ($= (G_F - P_T)\times1.1111$);
τ: [g/s/column] Mass transfer rate of isotope;
ϕ: [n/cm^2 s] Mean neutron flux of HMR ($= 5\times10^{13}$).

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