Assessment of tritium removal efficiency of WTRF with design and operating variables in the LPCE system

Si Woo Im,[a] Bum-Jin Chung,[a]
Do Hee Ahn,[b]

a Nuclear & Energy Engr. Dept. Cheju National Univ., 66 Jejudaehakno, Jeju-si, Jeju-do, Korea
b Korea Atomic Energy Research Institute 150 Duckjin-dong, Yusong-gu, Daejeon, Korea

Abstract

The internal tritium flow and hydrogen isotopic exchange reaction in a liquid phase catalytic exchange columns was modeled to investigate the effects of operating and design variables on the tritium extraction efficiency of the tritium removal facility in CANDU nuclear power plant. The selected design and operating variables are operating temperature, pressure, catalyst efficiency, number of theoretical stages in a hydrophilic bed, detritiation factor of the cryogenic distillation system and number of sections of the LPCE column. The value of the LPCE detritiation factor, which is defined by a ratio of tritium concentration in tritiated heavy water feed to that in heavy water product, is estimated 47 at a nominal operating condition, which is higher than 35 of performance requirement value by about 30%. The detritiation factor increases with increasing the design and operating variables except the operating pressure. The effect of temperature on the system performance is relatively strong, comparing with the other variables, because the temperature effects on the both of the separation factor and the molar flow rate of heavy water vapor in the LPCE column. The results of this study may be used as an operating reference for the LPCE system.

Introduction

Tritium is produced both by the nuclear fission and by neutron activation reactions in a nuclear power plant. The use of heavy water as moderator and coolant in Pressurized Heavy Water Reactors (PHWRs) permits greater burn up of natural uranium oxide fuel, but results in the production of tritium by neutron activation of the deuterium to a degree far in excess of that produced in fuel. The amount of tritium generated in PHWR by neutron capture reaction of deuterium in heavy water exceeds that in LWR by almost 100 times. The estimated equilibrium tritium activity in the coolant and moderator is 1.95 Ci/kg, 88.0 Ci/kg respectively. After 20 years operation of Wolsong unit 1, the tritium activity reached to 1.76 Ci/kg in coolant and 60.5 Ci/kg in moderator, by the end of 2003.

The presence of highly concentrated tritium in the heavy water systems of the CANDU plants, a typical PHWR, is a major source of radiation doses to the operating personnel and environment. For preventing the radiation hazards, the tritium reduction or removal in the systems had been suggested in early 1970s. Korea has 4 units of CANDU at Wolsong site; the tritium inventory will be reached to about 44.1 MCi by 2005. In order to reduce the tritium inventory, Wolsong tritium removal facility (WTRF) is now under construction with

completion date of the end of 2005. Tritium inventory will be decreased with the WTRF operation to about 10 MCi by 2013. The WTRF is designed to maintain the tritium activity in the moderator 10 Ci/kg. For the Wolsong unit 1, it will take about 4-5 years to reduce the tritium concentration under 10 Ci/kg the moderator. The amount of tritium removed is expected 2,576 atomic mole-T by 2013.

Tritium removal efficiency of the WTRF is surely depending on the design and operating variables, i.e. operating temperature and pressure, catalyst efficiency, number of stages a hydrophilic bed, detritiation factor of cryogenic distillation system, and number of sections within the Liquid Phase Catalytic Exchange (LPCE) system. Therefore, it is necessary to verify the relations for safe and reliable operation of the facility between those variables and the performance of the WTRF.

In this study, the tritium concentration profiles within the LPCE column was calculated to verify the LPCE system wouldmeet the designed tritium extraction efficiency. The change of detritiation factor of LPCE system was also simulated with the variations of the operating and design variables.

Modeling of the LPCE system

1. Overall description of reaction materials flow

The figure 1 shows a schematic diagram of hydrogen isotopic exchange process using Pt-catalyst with n sections arranged in a counter-current cascade. The LPCE system consists of two packed-bed columns, and a saturator is installed to humidify the deuterium gas at the bottom of column, and a condenser is installed to condensate the heavy water vapor at the top of column. Tritiated heavy water is fed to the top of column and the detritiated heavy water product is discharged at the bottom. The deuterium gas is entered from the cryogenic distillation system through the bottom section of column. The heavy water is flowing downward in contact with the up flowing vapor in a hydrophilic bed, and the vapor is up flowing with the deuterium gas in a catalyst bed.

In the n-th section, the tritium mole fraction of deuterium gas stream (x_n) and heavy water vapor stream (y_n) leaving a catalyst bed are

$$x_n = \left(1 - \frac{\eta_c \alpha_g}{\alpha_g + \gamma_g}\right) x_{n-1} + \left(\frac{\eta_c}{\alpha_g + \gamma_g}\right) y_{n-1}' \tag{1}$$

$$y_n = \left(\frac{\eta_c \alpha_g \gamma_g}{\alpha_g + \gamma_g}\right) x_{n-1} + \left(1 - \frac{\gamma_g \eta_c}{\alpha_g + \gamma_g}\right) y_{n-1}' \tag{2}$$

and the tritium mole fraction of heavy water feed stream (z_{n+1}) and vapor stream (y_n') leaving a hydrophilic bed,

$$z_{n+1} = \frac{[\alpha_v / \gamma_l - (\alpha_v / \gamma_l)^{Ns}] y_n - z_n / \gamma_l}{[\alpha_v / \gamma_l - (\alpha_v / \gamma_l)^{Ns}] / \alpha_v - 1 / \gamma_l} \tag{3}$$

$$y_n' = y_n - \frac{z_n - z_{n+1}}{\gamma_l} \tag{4}$$

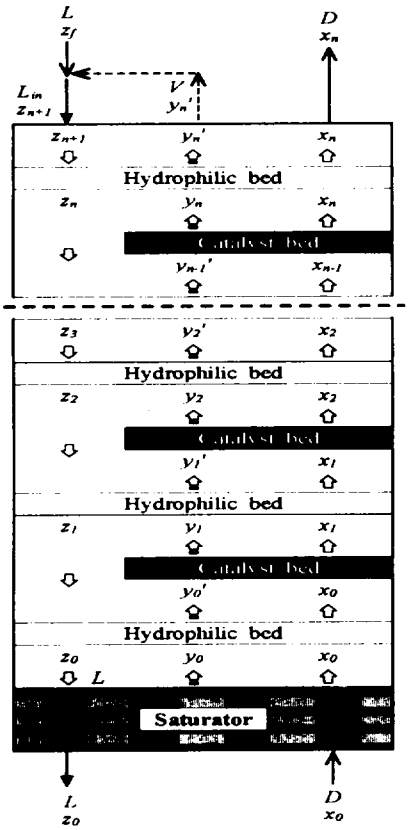

Fig. 1. A schematic representation of the LPCE process with n sections.

2.Numerical calculation of the internal tritium concentration in LPCE column

The detritiation factor of LPCE system and tritium concentrations x_i, y_i, z_{i+1}, y_i' in arbitrary section i can be calculated using the derived equations when the flow rate of heavy water feed, tritium mole fraction of feed water stream (z_f), the flow rate of deuterium stream, the detritiation factor of the cryogenic distillation process ($DF_{CD}=x_n/x_0$), the operating temperature (T), the operating pressure of the saturator (P_{sys}), efficiency of the catalyst bed, number of theoretical stages per hydrophilic bed, total number of section are given. The calculation procedures are summarized as follows,

a) Tritium mole fraction of heavy water product (z_0) and tritium mole fraction of entered deuterium gas (x_0) are assumed by using half interval method.

b) From the definition of the separation factor (α_v), tritium mole fraction of vapor stream y_0' is

Table 1. The summary of parameters and relations

Parameter	Symbol	Material balance equation
D_2O feed flow rate (mole/hr)	L	-
Internal D_2O feed flow rate (mole/hr)	L_{in}	-
D_2 flow rate (mole/hr)	D	-
Internal vapor flow rate (mole/hr)	V	-
Efficiency of a catalyst bed	η_c	$(x_{n-1}-x_n)/(x_{n-1}-x_{ne}$† $)$
Number of theoretical stages in a hydrophilic bed	Ns	$(y_n'-y_n)/(y_n'-y_{ne}$† $)$
Tritium separation factor between gas and vapor	α_g	$e^{(0.772-673.3/T+277052/T^2-3.1806E7/T^3+5.0868E8/T^4)}$‡
Tritium separation factor between vapor and liquid	α_v	$e^{(0.066-61.03/T+14198.4/T^2)}$
Molar flow rate ratio deuterium gas to vapor	γ_g	$D/V=(P_{sys}-P_{D2O})/P_{D2O}$*
Molar flow rate ratio vapor to water	γ_l	V/L_{in}

† x_{ne} and y_{ne} is equilibrium tritium concentration of each phase

‡ T is temperature of Kelvin degree

* $P_{D_2O}=10^{[7.01448-(1544.32+124209/T)/T]}$

$$y_0' = z_0 / \alpha_v \quad (5)$$

c) Tritium mole fraction of liquid stream leaving the first section z_1 is calculated using the material balance.

$$z_1 = \frac{L \times z_0 + V \times y_0'}{L_{in}} \quad (6)$$

d) Tritium concentrations x_1, y_1, z_2, y_1' are calculated using the derived equations. In a same way x_n, y_n, z_{n+1}, y_n' in section n are obtained in serial order.

Results and Discussions

A parametric study was performed using the computer program to investigate the effects of design and operating parameters on the detritiation factor of the LPCE system are investigated. The design and operating variables were selected in the process of establishing the model of the LPCE system, that need to be seriously considered in a point of view of system performance. The selected design and operating variables are operating temperature, pressure, catalyst efficiency, number of stages in a hydrophilic bed, detritiation factor of the cryogenic distillation system and number of sections of the LPCE column.

The nominal values and the ranges of the design and operating parameters varied in this study are summarized in Table 2. They are expected to cover fluctuation of variables, and certain installation problems of components or designing defects.

The figure 2 shows expected tritium profiles for the each reaction material in a nominal operation of the LPCE system.

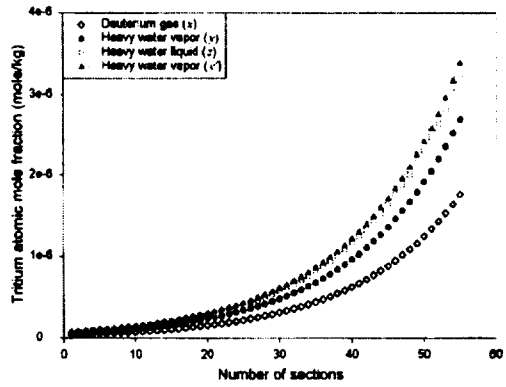

Fig. 2. Tritium concentration profiles of the each reaction material in the LPCE column. The mole flow rate of deuterium is double to the tritiated heavy water feed. The detritiation factor of LPCE system ($DF=z_1/z_0$) is calculated to about 47.

Table 2. Nominal values and variation range of parameters

Design and operating parameters	Nominal value	Variation range
LPCE column operating temperature	343 K = 70 ℃	55 ℃ < T < 75 ℃
LPCE column operating pressure	145 kPa	110 kPa < P < 200 kPa
Efficiency of a catalyst bed	0.9	0.6 < η_c < 1
Number of stages in a hydrophilic bed	2.0	1 < Ns < 3
Detritiation factor of the CD system	50	30 < DF_{CD} < 60
Number of sections in the LPCE columns	55	40 < $Nsec$ < 55

1. The effect of temperature and pressure

The temperature affects on the tritium separation factor and molar flow rate of vapor, and the pressure only affects on the molar flow rate of vapor. The increased temperature induces the decreasing of separation factor and increasing of internal heavy water vapor flow rate. Also, the vapor flow rate is increased with decreasing of pressure. They are activating the tritium transferring from the tritiated feed water to deuterium gas. Thus, the tritium separation factor (α) and molar flow rate ratio (γ) are affected by the variation of temperature and pressure.

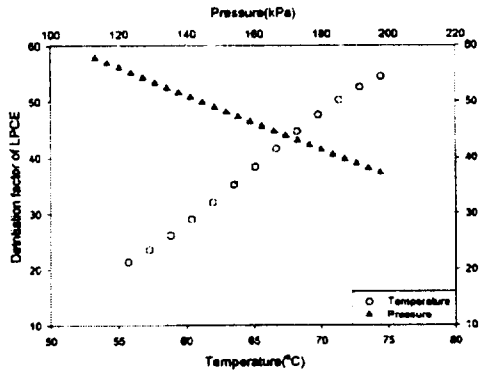

Fig. 3. The detritiation factor shows proportionality to the temperature, and inverse proportionality to the pressure. The curve fitting results for the ranges of variation are $DF(T)=1.85T-81.87$, $DF(P)=-0.233P+83.63$.

2. The effect of efficiency of catalyst bed and number of theoretical stages in a hydrophilic bed

The tritium is naturally exchanged between the vapor and the gas or between the vapor and the water by the difference of the volatility using the hydrogen isotopic exchange reaction. The catalyst activates the reaction; therefore, the high catalyst efficiency and large number of theoretical stages can transfer more tritium from the heavy water to the deuterium gas. The catalyst efficiency and number of theoretical stages are affected by the designing and installing the beds because they are closely related with the physical shape and quantity of catalysts.

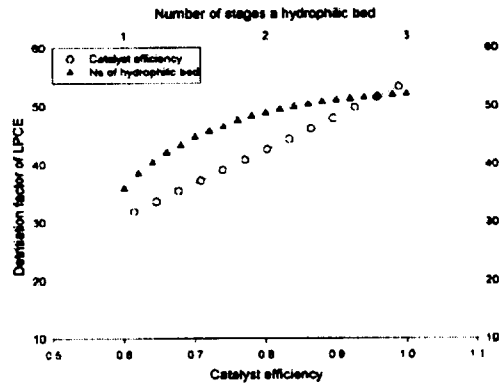

Fig. 4. The detritiation factor is increased with proportionally for catalyst efficiency and with the second order of a logarithmic function of the number of stages in a hydrophilic bed. The curve fitting results for the ranges of variation are $DF(\eta_c)=0.572\eta_c+30.476$, $DF(Ns)=-7.86\ [ln(Ns)]^2+23.14ln(Ns)+36.38$.

3. The effect of detritiation factor of the cryogenic distillation system and total number of section in the column

The initial tritium concentration of the supplied deuterium gas to the LPCE columns should be considered, since deuterium gas is a material for hydrogen isotopic exchange reaction. Supplying the low tritiated deuterium gas to the LPCE system is directly depending on the performance of the cryogenic distillation system.

The designed total number of sections in column consists of 55 sections. The performance of the system should be depending on that how many sections are installed.

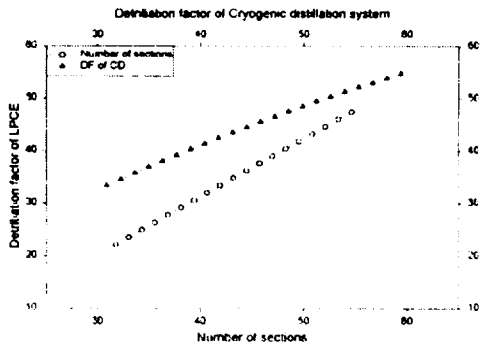

Fig. 5. The detritiation factor is increased linearly with the detritiation factor of cryogenic distillation system and total number of sections. The curve fitting results are for the range of variation are $DF(DF_{CD})=0.821DF_{CD}+6.49$, $DF(Nsec)=1.104Nsec-12.9$.

Conclusions

The LPCE system was proved that having a design margin to ensure the performance reliability to meet the WTRF design specification. The calculated detritiation factor of the WTRF LPCE system is 47 at a nominal condition, which is higher by about 30%, than 35, which is the requirement value.

The tritium extraction efficiency (detritiation factor) of the WTRF was varied in related with variations of the design and operating variables of the LPCE system. The variables to be worthy of attention suggested by author are operating temperature and pressure, because they are relatively easy to be changed and having effects on the almost all the components and parameters of the LPCE system such as catalyst, saturator, and vapor flow rate, separation factor. Especially, the operating temperature has relatively strong effects because it affects both the tritium separation factor and molar flow rate ratio.

These results can be used as an operating reference for the LPCE system. Also, the operating nominal condition can be newly setting up to maximize the detritiation factor of the WTRF under the safe operating the facility based upon the results.

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