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Analysis of Heavy Water Separation Cascade Using Bithermal H_2/H_2O Exchange Process

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Abstract

The 3-stage cascade composed of the multisection-type bithermal H_2/H_2O-exchange columns was suggested for heavy water separation. In order to study the separation characteristics for the cascade, a matrix equation with 18 simultaneous equations was composed and the concentrations and flow rates were calculated for the all parts of the cascade. Product D-concentration decreases and extraction yield increases with increasing cut in each stage, which is one of the principal parameters of the separation characteristics. The optimization of the 3-stage cascade can be made by case study using the matrix equation.

I. Introduction

In recent years, a hydrophobic Pt catalyst for the water/hydrogen exchange was invented and developed by the AECL scientists and the studies on the countercurrent liquid water/hydrogen exchange process using the trickle bed system were reported by the AECL engineers[1]. On the Pt surface happens the isotopic exchange reaction between water vapor and hydrogen due to the hydrophobic Pt catalyst and then the isotopic exchange reaction between liquid water and water vapor takes place at the interface between liquid water and water vapor. As the interface area is too small in case of using only the hydrophobic Pt catalyst bed, the AECL researchers used the mixed bed of hydrophobic Pt catalyst and hydrophilic inert particles for the purpose of increasing the water/vapor contact surface area.

The new multisection-type H_2/H_2O-exchange column without the superheating section was developed by the Institute of Physical and Chemical Research [2,3] in Japan. Heavy water separation characteristics was studied numerically for a pair of dual temperature multisection-type H_2/H_2O-exchange columns at normal pressure packed with the hydrophobic Pt catalyst and Sulzer CY-Packing[4]. The influence of the operating pressure and temperature on the specific column volume was studied for the design of the system[5].

In this paper the authors show the basic theory for the design of 3-stage heavy water separation cascade composed of the multisection-type bithermal H_2/H_2O-isotopic exchange columns using the hydrophobic Pt catalyst.

II. Description of Process

The schematic diagram of 3-stage heavy water separation cascade composed of the multisection-type bithermal H_2/H_2O-isotopic exchange column is shown in Fig. 1.

The main natural water is fed to the top of the cold exchange column of the 1st stage and the depleted water is drawn from the bottom of the hot exchange column of the same stage. The hydrogen is circulated between the cold and hot exchange columns, extracting the deuterium from the countercurrently flowing water in the exchange column of the 1st stage.

In the exchange columns of the 2nd stage, the hydrogen drawn from the exchange column of the 1st stage is fed to the bottom of the hot exchange column of the 2nd stage and flows up through the hot and cold exchange columns and then is returned to the 1st stage. The water is circulated between the cold and hot exchange columns countercurrently to the hydrogen as a deuterium transfer medium.

In the exchange column of the 3rd stage, the hydrogen drawn from the exchange column of the 2nd stage is fed to the bottom of the hot exchange column of the 3rd stage and flows up through the hot and cold exchange columns and then is returned to the 2nd stage. The water is circulated between the cold and hot exchange columns countercurrently to the hydrogen, extracting the deuterium from the hydrogen stream. The enriched water is drawn from the bottom of the cold exchange column and then fed to the final enrichment stage. At the same time, the auxiliary natural water is fed to the top of the hot exchange column of the 3rd stage in the same quantity as that of the enriched water drawn from the 3rd stage.

In each exchange column, there are alternatively hydrophobic Pt catalyst bed for catalytic reaction section and packed bed or perforated plates for scrubbing section and heat exchanger with a function of humidification at the bottom as shown in the previous papers[4,5]. There is also a vapor/liquid separator with a function of heat exchange between the cold and hot exchange columns.

Ⅲ. Analysis for Multisection-Type H_2/H_2O-Isotopic Exchange Column Cascade

The numerical analysis for the design of 3-stage cascade composed of the above-mentioned pair of multisection-type bithermal exchange columns shown in this paper is based on the analytical expressions derived from the finite difference equation shown in the previous papers[4,5].

Assuming at first that the D-mole fraction is less than 0.20, we find that the separation factor for catalytic exchange α_g is nearly the ratio of D-mole fraction of water vapor to that of hydrogen and the separation factor α_l is also nearly the ratio of D-mole fraction of liquid water to that of water vapor.

Then the catalytic exchange efficiency η_c and the scrubbing efficiency η_b are defined as follows:

$$\eta_c=\frac{x_{n-1}^H-x_n^H}{x_{n-1}^H-(x_n^H)_e},\quad \eta_b=\frac{x_n^W-y_n^W}{x_n^W-(y_n^W)_e} \tag{1}$$

From the results written in the papers by Shimizu[4,5], the following expressions are written :

$$\begin{aligned}
x_n^H&=A\ x_{n-1}^H+B\ y_{n-1}^w\\
x_n^W&=C\ x_{n-1}^H+D\ y_{n-1}^W\\
y_n^W&=E\ z_n^W+F\ x_{n-1}^W\\
z_{n+1}^W&=G\ z_n^W+H\ x_n^W \quad (\mathrm{n}:\ \mathrm{Integer}\ \geq\ 1)
\end{aligned} \tag{2}$$

where $A=(1-\frac{\alpha_g\eta_c}{\alpha_g+\gamma_g})$, $B=\frac{\eta_c}{\alpha_g+\gamma_g}$, $C=\frac{\alpha_g\gamma_g\eta_c}{\alpha_g+\gamma_g}$, $D=(1-\frac{\gamma_g\eta_c}{\alpha_g+\gamma_g})$

$E=\frac{\eta_b}{\alpha_l-\gamma_l\eta_b}$, $F=(1-\eta_b)-\frac{\gamma_l\eta_b^2}{\alpha_l-\gamma_l\eta_b}$, $G=\frac{\alpha_l}{\alpha_l-\gamma_l\eta_b}$, $H=\frac{-\alpha_l\gamma_l\eta_b}{\alpha_l-\gamma_l\eta_b}$

$\gamma_g=\frac{N_G^H}{N_G^W}=\frac{\pi-P_{H_2O}}{P_{H_2O}}$, $\gamma_l=\frac{N_G^W}{N_L^W}$

From Eq.(2), the following finite difference equation concerning x^H can be derived :

$$x_{n+3}^H+px_{n+2}^H+qx_{n+1}^H+\gamma x_n^H=0 \tag{3}$$

where $p=-(A+DF+G)$, $q=(ADF-BCF+AG+DFG-DEH)$

$\gamma=(BC-AD)(FG-EH)$

The solution becomes as follows :

$$x_n^H=K_1g_1^n+K_2g_2^n+K_3g_3^n \quad (\mathrm{n}\geq 0) \tag{4}$$

where K_1, K_2, K_3 are constants and g_1, g_2, g_3 are real roots of the following characteristic equation: $g^3+pg^2+qg+\gamma=0$ $(\mathrm{g}\neq 0)$

For the 1st stage of the 3-stage bithermal H_2/H_2O-exchange cascade the following expressions can be derived concerning the deuterium concentration :

For the cold column,

$$x_n^H(1)=K_1(1)g_1^n(1)+K_2(1)g_2^n(1)+K_3(1)g_3^n(1) \tag{5}$$

$$x_n^H(1)=K_1(1)[C(1)+\frac{D(1)}{B(1)}(g_1(1)-A(1))]g_1^{n-1}(1)$$

$$+K_2(1)[C(1)+\frac{D(1)}{B(1)}(g_2(1)-A(1))]g_2^{n-1}(1)$$

$$+K_3(1)[C(1)+\frac{D(1)}{B(1)}(g_3(1)-A(1))]g_3^{n-1}(1),\ (n\geq 1) \tag{6}$$

$$y_n^W(1)=K_1(1)(\frac{g_1(1)-A(1)}{B(1)})g_1^n(1)+K_2(1)(\frac{g_2(1)-A(1)}{B(1)})g_2^n(1)+K_3(1)(\frac{g_3(1)-A(1)}{B(1)})g_3^n(1),(n\geq 0) \tag{7}$$

$$Z_n^W(1)=K_1(1)\frac{1}{B(1)E(1)}[(g_1(1)-A(1))g_1(1)-F(1)(B(1)C(1)+D(1)(g_1(1)-A(1))]g_1^{n-1}(1)$$

$$+K_2(1)\frac{1}{B(1)E(1)}[(g_2(1)-A(1))g_2(1)-F(1)(B(1)C(1)+D(1)(g_2(1)-A(1))]g_2^{n-1}(1)$$

$$+K_3(1)\frac{1}{B(1)E(1)}[(g_3(1)-A(1))g_3(1)-F(1)(B(1)C(1)+D(1)(g_3(1)-A(1))]g_3^{n-1}(1),(n\geq 1) \tag{8}$$

For the hot column, the similar derivation can be made concerning the hot column. In this case, a bar must be attached to each symbol and Eqs. (9)~(12) corresponding to Eqs. (5)~(8) can be expressed as an example of the following equation (9).

$$\overline{x_n^H}(1)=\overline{K_1}(1)\overline{g_1^n}(1)+\overline{K_2}(1)\overline{g_2^n}(1)+\overline{K_3}(1)\overline{g_3^n}(1),\ (n\geq 0) \tag{9}$$

For the 2nd stage of the 30 stage bithermal H_2/H_2O-exchange process the similar treatments can be made. In this case, (1) of each symbol in Eqs. (5)~(12) must be replaced by (2). For example,

$$x_n^H(2)=K_1(2)g_1^n(2)+K_2(2)g_2^n(2)+K_3(2)g_3^n(2) \tag{13}$$

For the 3rd stage of the 3-stage bithermal H_2/H_2O-exchange process the similar treatments can be made too. In this case, (2) of each symbol in Eq. (13) must be replaced by (3). For example,

$$x_n^H(3)=K_1(3)g_1^n(3)+K_2(3)g_2^n(3)+K_3(3)g_3^n(3) \tag{14}$$

The constants

$$K_1(1), K_2(1), K_3(1), \overline{K_1}(1), \overline{K_2}(1), \overline{K_3}(1);$$
$$K_1(2), K_2(2), K_3(1), \overline{K_1}(2), \overline{K_2}(2), \overline{K_3}(2);$$
$$K_1(3), K_2(3), K_3(3), \overline{K_1}(3), \overline{K_2}(3), \overline{K_3}(3);$$

can be obtained from the 18 boundary conditions.

Putting Eqs. (5)~(14) and the flow relations into the 18 boundary conditions, we get a matrix equation composed of 18 simultaneous simple equations.

In order to get the concentrations and flow rates for each part of the cascade, the values of the operating conditions must be at first put into the matrix equation.

Ⅳ. Results and Discussion

The concentrations and flow rates for all parts of the 3-stage heavy water separation cascade were determined by solving the matrix equation. Figures 3 and 4 show D-concentration of product stream and extraction yield as a function of cut in each stage under the following conditions :

M_1/N_1 = 145/95, M_2/N_2 = 295/145, M_3/N_3 = 395/145, π = 30kg/㎠

$T_c = T_r = 90℃$, $T_h = 205℃$, $\eta_c = \eta_b = 0.9$, $\gamma = 2.05$, $\theta_1 = \theta_2$
Product D-concentration decreases and extraction yield increases with increasing cut in each stage, which is one of the principal parameters of the separation characteristics.

V. Conclusion

Through the analysis written in this paper for the 3-stage heavy water separation cascade composed of the multisection-type bithermal H_2/H_2O-exchange columns, the concentrations and flow rates can be computed for the all parts of the cascade. Product D-concentration decreases and extraction yield increases with increasing cut in each stage, which is one of the principal parameters of the separation characteristics. The optimization of the 3-stage cascade can be made by case study using the matrix equation.

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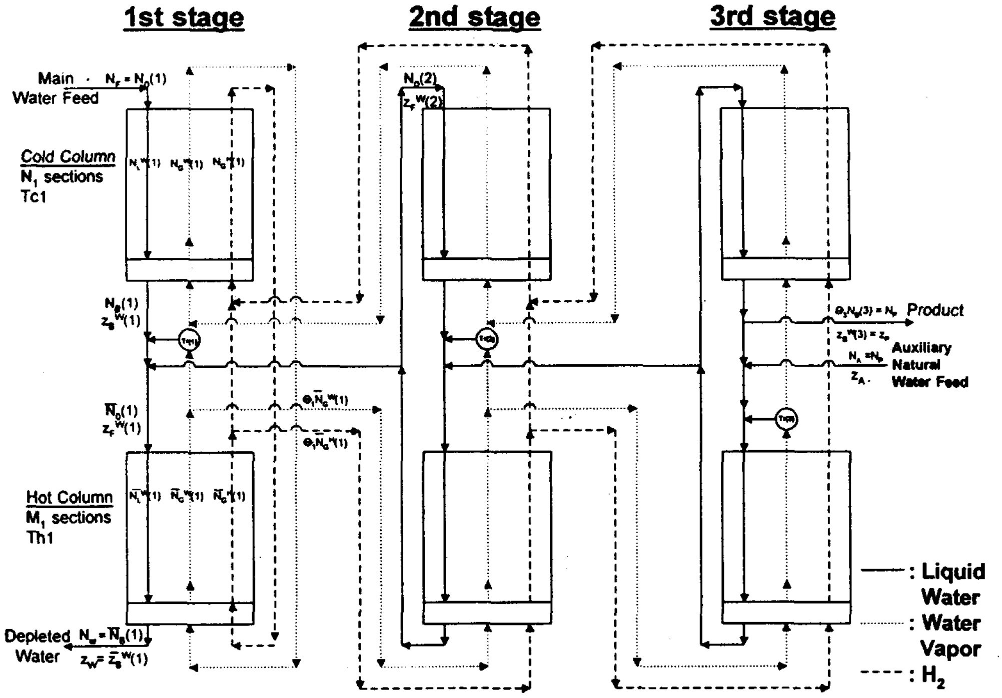

Fig. 1. Schematic diagram of the 3-stage heavy water separation cascade composed of the multisection-type bithermal H_2/H_2O-exchange columns

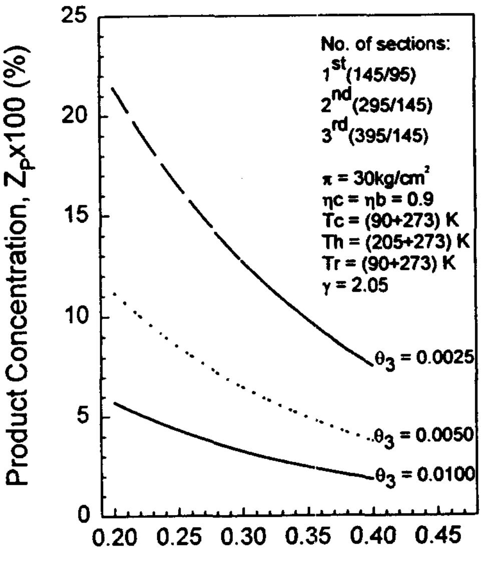

Fig. 2. D-concentration in product stream as a function of cut in each stage

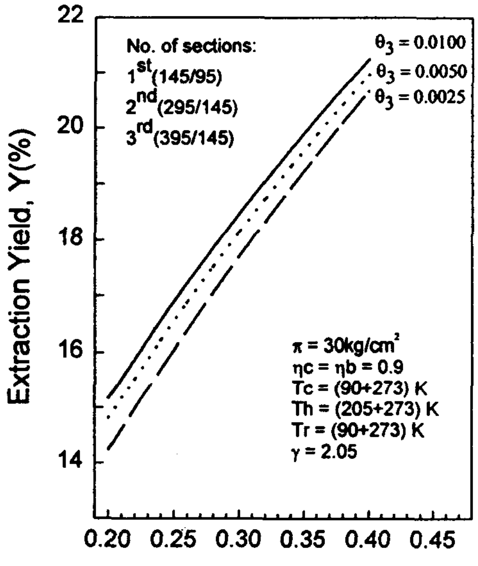

Fig. 3. Extraction yield as a function of cut in each stage