Analele Universităţii de Vest din Timişoara
Vol. XLIX, 2006
Seria Fizică

COMPUTER SIMULATION OF HYDROGEN ISOTOPES SEPARATION PROCESS BY SIMPLE CRYOGENIC DISTILLATION

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Abstract
In the last fifty years many studies have proved that the one of the best process of tritium elimination is the hydrogen isotopes separation by cryogenic distillation. On the other hand this process is now recommended (from economical and technical reasons) in production of tritium used like fuel in the next fusion reactors. A computer code is developed for exact simulation based on the present models of cryogenic distillation column, modified numerical methods (for ensuring the convergence of algorithms) and experimental data.
Keywords: hydrogen, isotope, cryogenic, distillation, simulation.

1. Introduction

Many studies have proved that the one of the best process of tritium elimination is the hydrogen isotopes separation by cryogenic distillation. This process has numerous advantages - large separation factors, comparatively large processing flow rate even with compact scale devices, relatively small power consumption, negligible tritium permeation (because of extremely low operating temperature) - whereas it has a few disadvantages - comparatively large tritium inventory, necessity of safeguard against the loss of refrigerant etc.

The design and implementation of column distillation method for hydrogen isotopes require both experimental studies and computer simulations. Particularly, the computer simulation is indispensable for design and performances precognition of cryogenic distillation column or cryogenic distillation column cascade. In this way the present study provides a great help to development of simulation models and simulation procedures of hydrogen isotopes cryogenic distillation column.

2. Method and samples

In present they are several calculation procedures of multicomponent distillation developed for hydrocarbure processing with large separation factors. They require same critique modifications or improvements in several aspects to be applied to hydrogen isotope separation by cryogenic distillation with small separation factors. The column model for

mathematical simulation is shown in Fig. 1 (the first stage is the condenser and the N-stage is the reboiler).

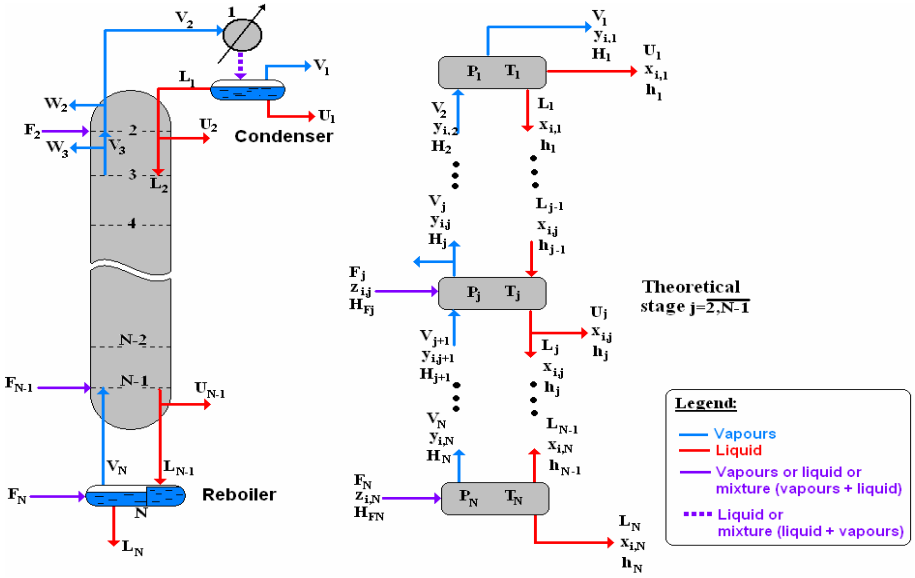

Fig. 1. Principal scheme of classical distillation column

The basic equations are derived from the requirements of conservation and for phase equilibrium on each stage:

- component material balances

$$\begin{aligned} &V_2 y_{i,2} - V_1 y_{i,1} - (L_1 + U_1) x_{i,1} = 0 \\ &L_{j-1} x_{i,j-1} + V_{j+1} y_{i,j+1} + F_j z_{i,j} - (V_j + W_j) y_{i,j} - (L_j + U_j) x_{i,j} = 0, \qquad j = \overline{2, N-1} \\ &L_{N-1} x_{i,N-1} + F_N z_{i,N} - V_N y_{i,N} - L_N x_{i,N} = 0 \end{aligned} \tag{1}$$

- heat balances

$$E_{j,in} = L_{j-1} h_{j-1} + V_{j+1} H_{j+1} + F_j H_{Fj} + H_{Lj} (x_{3,j} + x_{5,j} + 2x_{6,j}) a \frac{\lambda}{2} + q_j$$

$$E_{j,out} = (L_j + U_j) h_j + (V_j + W_j) H_j + Q_j; \quad E_j = 1 - \frac{E_{j,out}}{E_{j,in}} = 0, \qquad j = \overline{2, N-1} \tag{2}$$

- vapor-liquid equilibrium

$$y_{i,j} = K_{i,j} x_{i,j}, \quad K_{i,j} = \xi_{i,j} \frac{p^o_{i,j}}{P_j} \tag{3}$$

- global material balance:

$$L_j + U_1 + V_1 - V_{j+1} - \sum_{k=2}^{j} (F_k - W_k - U_k) = 0, \qquad j = \overline{2, N-1} \tag{4}$$

Finally we are obtained one tridiagonal matrix equation:

$$\begin{bmatrix} B_1 & C_1 & 0 & \cdots & 0 \\ A_2 & B_2 & C_2 & \cdots & 0 \\ \vdots & & & & \vdots \\ 0 & \cdots & A_{N-1} & B_{N-1} & C_{N-1} \\ 0 & \cdots & 0 & A_N & B_N \end{bmatrix} \cdot \begin{bmatrix} x_{i,1} \\ x_{i,2} \\ \vdots \\ x_{i,N-1} \\ x_{i,N} \end{bmatrix} = \begin{bmatrix} D_{i,1} \\ D_{i,2} \\ \vdots \\ D_{i,N-1} \\ D_{i,N} \end{bmatrix} \qquad i = \overline{1, m} \tag{5}$$

where:

$$B_1 = -(L_1 + V_1 K_{i,1} + U_1), \qquad C_1 = V_2 K_{i,2}, \qquad D_1 = 0,$$
$$A_j = L_{j-1}, \qquad B_j = -(V_j + W_j) K_{i,j} - (L_j + U_j), \qquad C_j = V_{j+1} K_{i,j+1}, \qquad D_j = -F_j z_{i,j}, \qquad j = \overline{2, N-1} \qquad (6)$$
$$A_N = L_{N-1}, \qquad B_N = -V_N K_{i,N} - L_N, \qquad D_N = -F_N z_{i,N}$$

The convergence technique at each iterative step is very important in solving the nonlinear simultaneous equations. Usually it is used the multidimensional Newton-Raphson method. According to our tries the achievement of convergence is apt to become more difficult as number of theoretical stages and reflux ratio increase. Strict separation requirements are also one of the reasons for difficulty of the cryogenic distillation calculation.

In the part of our simulation program dedicated of simple cryogenic distillation column a personal technique based on inverse matrix method are used to ensure stability.

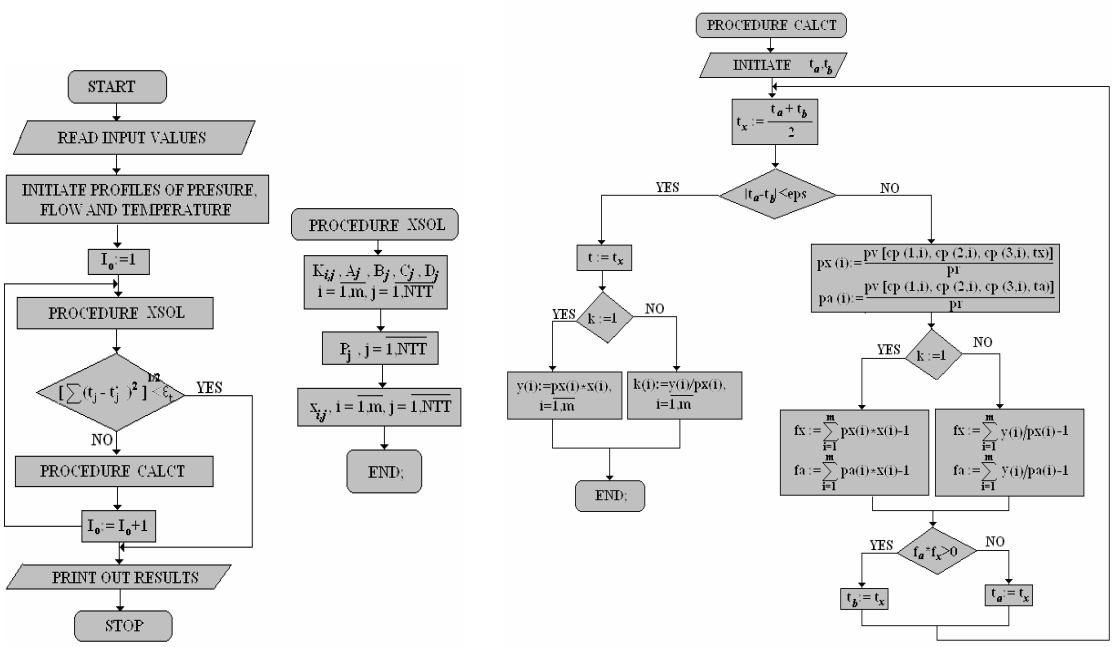

Fig. 2. Calculation algorithm for simple cryogenic distillation column

3. Results and Discussions

The reference conditions given completely in Table 1 have been assumed initially based on experimental results or theoretical considerations, respectively. Feed location (N_F) is fixed to be *N/2*. The vapors pressure values have been estimated using experimental values of *Mittelhauser* [4] for critical point of hydrogen, deuterium and tritium, and their diatomic combination and the interpolation program JANDEL SCIENTIFIC (LOTUS) (*ncc* = 3 - the number of approximate constants for each components). The estimation of vapor pressure at

each theoretical stage has been made by assuming the linear dependence pressure-temperature (constants k_a and k_b).

Table 1. Reference condition of simple column assumed for parametric studies

Number of components (nc)		6
Number of total theoretical stages (ntt)		30
Separator pressure		2.247,24 Torr
Vapor pressure		2.247,49 Torr
Pressure drop		0,25 Torr
Number of approximate constants (ncc)		3
Flow rate of feed stream		100 g-mol/h
Composition of feed stream	H_2	$0{,}13539 \cdot 10^{-3}$
	HD	$0{,}10437 \cdot 10^{-1}$
	HT	$0{,}92920 \cdot 10^{-2}$
	D_2	0,24757
	DT	0,48443
	T_2	0,24814
Approximate constant k_a		0,00266
Approximate constant k_b		18,68400
Feed location (ntf)		15
Condenser		total condenser (v[1] = 0)
Reflux ratio (l[1])		10 g-mol/h
Flow rate of top stream (D)		0 g-mol/h
Flow rate of bottom stream (W)		100 g-mol/h
Temperature of feed stream		24,67 K

4. Conclusions

The simulation procedure of simple cryogenic distillation column has tested for the variation of input variables. This procedure is applicable and effective in such cases that the cut of mixture of each stage is prescribed and require to be independent of concentration; the stage separation factors are given as input variables in large limits.

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