

JAERI - M  
93-188

SEPARATION CHARACTERISTICS OF CRYOGENIC DISTILLATION  
COLUMN HAVING FEEDBACK STREAM  
—SEPARATION EXPERIMENT WITH H-D SYSTEM—

October 1993

Toshihiko YAMANISHI, Takumi HAYASHI, Hirofumi NAKAMURA  
Kenji OKUNO, R. H. SHERMAN\*, D. TAYLOR\*, J. W. BARNES\*  
and J. R. BARTLIT\*

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編集兼発行 日本原子力研究所  
印刷 山田軽印刷所

Separation Characteristics of Cryogenic Distillation Column  
Having Feedback Stream  
- Separation Experiment with H-D System -

Toshihiko YAMANISHI, Takumi HAYASHI, Hirofumi NAKAMURA, Kenji OKUNO  
R.H. SHERMAN<sup>\*</sup>, D. TAYLOR<sup>\*</sup>, J.W. BARNES<sup>\*</sup> and J.R. BARTLIT<sup>\*</sup>

Department of Fusion Engineering Research  
Naka Fusion Research Establishment  
Japan Atomic Energy Research Institute  
Naka-machi, Naka-gun, Ibaraki-ken

(Received September 1, 1993)

The first experiment for the column having a feedback stream was successfully performed with H-D system at the TSTA. Several computer codes were arranged for simulation of this column. Slight modifications were made on the computer codes to obtain better convergence in the simulation and to consider more factors such as dead volumes and by-pass flows. Characteristics of the column having a feedback stream were studied by the simulation works with the H-D system. It was observed that flow rate and location of side stream had considerable effects on column performance. A method for determination of the flow rate and location of side stream was also discussed from the simulation results. A significant result observed from the experiment was that high purity D<sub>2</sub> was obtained from bottom of the column having a feedback stream; while for the column without the feedback stream, HD was a major component in the bottom stream. The effect of the feedback stream was thus experimentally verified.

Keywords: Cryogenic Distillation, Feedback Stream, Breeder Blanket,  
Hydrogen Isotopes, Computer Simulation, Side Stream, Column  
Performance, TSTA

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\* Los Alamos National Laboratory

フィードバック流れを持つ深冷蒸留塔の分離特性  
－H－D系による分離実験－

日本原子力研究所那珂研究所核融合工学部

山西 敏彦・林 巧・中村 博文・奥野 健二

R.H.SHERMAN\*・D.TAYLOR\*

J.W.BARNES\*・J.R.BARTLIT\*

(1993年9月1日受理)

フィードバック流れを持つ深冷蒸留塔の最初の実験が、H－D系で成功裏に行なわれた。この塔のシミュレーションのために幾つかの計算コードを整備し、より良い収束特性を得、塔内のバイパス流あるいはよどみ等のファクターを考慮できるように、コードに適当な修正を加えた。これらコードを用いて、H－D系で、フィードバック流れを持つ塔の特性をシミュレーションにより検討した。その結果、抜き出し流の流量及びその抜き出し箇所が、塔性能に最も大きな影響を持つことを確認した。更に、抜き出し流量及び抜き出し箇所の決定法を検討した。実験によって認められた1つの重要な結果は、フィードバック流れを持つ塔により、高純度のD<sub>2</sub>が塔底より得られたことである。これに対しフィードバックを持たない塔では塔底の主成分はHDであり、対照的な結果を示した。このように、実験によりフィードバック流れの効果を実証することができた。

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## 1. Introduction

Hydrogen isotope separation by cryogenic distillation is expected to be best suited to the needs in the fuel circulation system of fusion reactors: a comparatively large flow rate can be processed even with a compact scale; and it has relatively high separation performance. The separation characteristics of the cryogenic distillation columns have been studied by the authors for bench scale columns<sup>(1)~(3)</sup>. The HETP values were measured; and its dependence on the vapor velocity within the column and the reflux ratio was discussed in detail. The experiments by actual scale columns have also been carried out at the TSTA (Tritium Systems Test Assembly)<sup>(4)~(6)</sup>. A significant conclusion was that the engineering data for bench scale columns can directly be applied to the actual scale columns. A great deal of valuable information for the design the cryogenic distillation columns were thus obtained.

In the main fuel cycle of the fusion reactor, an isotope mixture of the six molecular species is separated into four streams:  $H_2$ -HD,  $D_2$ , DT and  $T_2$ . In the breeder blanket interface (BBI), a mixture of  $H_2$ , HT and  $T_2$  is separated into two streams of  $H_2$  and  $T_2$ . In these hydrogen isotope separations, a major concern must be made how do we treat the molecular specie of HT. Hydrogen must be removed as an impurity, while tritium must be recovered. It is already known that H and T can be separated by a single cryogenic distillation column having a feedback stream. The molecular specie of HT was decomposed to  $H_2$  and  $T_2$  through an equilibrator. Kinoshita et al. have developed computer simulation codes for this column; and have studied the separation characteristics<sup>(7),(8)</sup>. They proposed a two-column cascade for the main fuel cycle system<sup>(9)</sup>, which is composed of two columns having feedback streams. For the BBI, a two-column cascade, which is composed of one ordinary column and one column having a feedback stream, was proposed<sup>(10)</sup>. From their simulation studies, the column having the feedback stream was recognized to be considerably useful. However, the effect of the

feedback stream on separation performance of the column has not been still exactly understood. Another important problem is that there have been no report that experimentally verifies the effect of the feedback stream. To overcome these problems, modification was made on the ISS (Isotope Separation System) at the TSTA: a feedback stream, which can be withdrawn from several locations, was installed on one of the columns. Experiments were initiated with H-D and D-T systems from April 1993 to obtain data for the column having the feedback stream.

The present paper reports the first experiment for the column having a feedback stream with the H-D system. The computer codes for simulation of the column are first introduced. The effect of feedback stream and a method for determination of column parameters are discussed by the simulation results. The test plan and test results of the experiment are then briefly reported. The detailed experimental results will be published in the proceeding of 15th Symposium on Fusion Engineering.

## 2. Mathematical Simulation Procedure

Five computer simulation codes are used in the present paper. The detailed mathematical simulation procedures are described in Ref. 11 and 12, so that summaries of these codes are introduced in this chapter. The important points in the simulation codes, which should be paid attention to get convergence in the simulation, are also proposed.

### 2.1 Steady-state simulation for ordinary column

In accordance with the stage model, basic equations are derived as follows.

$$\begin{bmatrix} B_{i,1} & C_{i,1} & 0 & \dots & \dots \\ A_{i,2} & B_{i,2} & C_{i,2} & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots \\ & A_{i,j} & B_{i,j} & C_{i,j} & \\ 0 & \dots & \dots & A_{i,N} & B_{i,N} \end{bmatrix} \cdot \begin{bmatrix} x_{i,1} \\ x_{i,2} \\ \vdots \\ x_{i,j} \\ x_{i,N} \end{bmatrix} = \begin{bmatrix} D_{i,1} \\ D_{i,2} \\ \vdots \\ D_{i,j} \\ D_{i,N} \end{bmatrix}, \quad (1)$$



feedback stream on separation performance of the column has not been still exactly understood. Another important problem is that there have been no report that experimentally verifies the effect of the feedback stream. To overcome these problems, modification was made on the ISS (Isotope Separation System) at the TSTA: a feedback stream, which can be withdrawn from several locations, was installed on one of the columns. Experiments were initiated with H-D and D-T systems from April 1993 to obtain data for the column having the feedback stream.

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where

$$A_{i,j} = L_{j-1}, \quad B_{i,j} = -V_j K_{i,j} - L_j, \quad C_{i,j} = V_{j+1} K_{i,j+1}, \quad D_{i,j} = -F_j z_{i,j}.$$

Kinoshita has proposed the Newton-Raphson method to solve Eq. (1)<sup>(11)</sup>. However, the Newton-Raphson method does not give us convergence in the case where initial values of independent variables, which are the temperatures on the stages for this case, are not properly set. On the other hand, the tri-diagonal method<sup>(12)</sup> gives the solution of this equation except the case where the reflux ratio of the column is extremely large. In addition, this method can easily consider all the factors such as the differences in latent heat of vaporization among the hydrogen isotopes; decay heat of tritium; and nonideality of hydrogen isotope solutions. For the above reasons, we used the tri-diagonal method in the present report. If it is difficult to obtain the solution by the tri-diagonal method, the following procedure would be effective to get the solution.

- (i) Usually, the solution can be obtained by the tri-diagonal method in 50~100 times of iteration.
- (ii) End the calculation by the tri-diagonal method in the case where the iteration exceeds ~200 times.
- (iii) Carry out the simulation by the Newton-Raphson method. The calculated results by the tri-diagonal method after ~200 times of iteration are used as initial values.

## 2.2 Dynamic simulation

It is expected that qualitative aspects of the conclusions are not vitiated at all even if the calculation is made by using a simplified model neglecting the factors described above. Basic equations are expressed by

$$H_{Lj} \frac{dx_{i,j}}{dt} = V_{j+1} y_{i,j+1} + L_{j-1} x_{i,j-1} - L_j x_{i,j} - V_j y_{i,j} + F_j z_{i,j}. \quad (2)$$

Equation (2) can be solved by applying Ballard-Brosilow's algorithm<sup>(11)</sup>. For the case where the column has a feedback stream, the equation is rewritten as follows:

$$H_{Lj} \frac{dx_{i,j}}{dt} = V_{j+1}y_{i,j+1} + L_{j-1}x_{i,j-1} - L_jx_{i,j} - (V_j + W_j)y_{i,j} + F_j^{\text{ex}}z_{i,j}^{\text{ex}} + F_j^{\text{in}}z_{i,j}^{\text{in}} \quad (3)$$

The internal feed stream is an equilibrium state at room temperature, whose composition,  $z_{i,j}^{\text{in}}$ , is calculated from the atom fraction of the side stream. Sometimes, the factors such as the vapor holdup in the condenser, by-pass flow, and dead liquids in the column are not negligible for the simulation of dynamic behavior of the column. For this case, we consider these factors by the computer code using the Improved Euler method<sup>(11),(12)</sup> because of its simple algorithm.

### 2.3 Steady-state simulation for the column having a feedback stream

If the atom fraction of protium in the side stream,  $Z_H$ , and that of deuterium,  $Z_D$ , are given, the composition and temperature of the stream fed to the column can be determined. Then, the composition and temperature distributions within the column are computed by the main distillation calculation. The computed values of the atom fraction of protium and deuterium in the side stream,  $Z_H$  and  $Z_D$ , must be equal to  $Z_H$  and  $Z_D$ , respectively. This means solving the following nonlinear simultaneous equations:

$$f_H(Z_H, Z_D) = Z'_H - Z_H, \quad f_D(Z_H, Z_D) = Z'_D - Z_D \quad (4)$$

Equation 4 can be solved by the two-dimensional Newton-Raphson method<sup>(8)</sup>:

$$\left. \begin{aligned} Z_H^{i+1} &= Z_H^i - \{f_H(\partial f_D / \partial Z_D) - f_D(\partial f_H / \partial Z_D)\} / E \\ Z_D^{i+1} &= Z_D^i - \{f_D(\partial f_H / \partial Z_H) - f_H(\partial f_D / \partial Z_H)\} / E \\ E &= (\partial f_H / \partial Z_H)(\partial f_D / \partial Z_D) - (\partial f_D / \partial Z_H)(\partial f_H / \partial Z_D) \end{aligned} \right\}, \quad (5)$$

where  $i$  denotes the iteration number. The partial derivative has been numerically calculated by using the following approximation<sup>(8)</sup>:

$$(\partial f_j / \partial Z_k) \cong \frac{f_j(Z_k + \Delta Z_k) - f_j(Z_k)}{\Delta Z_k} \quad (6)$$

However, it was observed that slight modification of this equation significantly quickens the convergence when Eq. (4) was solved by use of Eq. (5):

$$(\partial f_j / \partial Z_k) \cong \frac{f_j(Z_k + \Delta Z_k) - f_j(Z_k - \Delta Z_k)}{2\Delta Z_k} \quad (7)$$

The main distillation calculation is performed by finding out the solutions of the following  $N$ -dimensional nonlinear simultaneous equations<sup>(12)</sup>:

$$\left. \begin{aligned} S_1(T_1, \dots, T_N) &= 0 \\ \dots \\ S_N(T_1, \dots, T_N) &= 0 \end{aligned} \right\}, \quad (8)$$

where  $S_j$  is defined by

$$S_j = \sum_{i=1}^m K_{i,j} x_{i,j} - 1 \quad (9)$$

Equation (8) is solved by application of the modified Newton-Raphson method<sup>(11)</sup>. The new values of the independent variables for the  $(\beta+1)$ 'th iteration are calculated from

$$T_j^{\beta+1} = T_j^{\beta} + \text{sign}(\alpha, \Delta T_j^{\beta}) \quad \text{or} \quad T_j^{\beta+1} = T_j^{\beta} + \Delta T_j^{\beta} \quad , \quad (10)$$

where

$$\begin{bmatrix} \partial S_1 / \partial T_1 & \dots & \partial S_1 / \partial T_N \\ \dots & \dots & \dots \\ \partial S_N / \partial T_1 & \dots & \partial S_N / \partial T_N \end{bmatrix} \begin{bmatrix} \Delta T_1^{\beta} \\ \dots \\ \Delta T_N^{\beta} \end{bmatrix} = - \begin{bmatrix} S_1 \\ \dots \\ S_N \end{bmatrix} \quad (11)$$

The nonideality of the solution and the heat balance are considered in an outer iterative loop. In Kinoshita's report<sup>(7)</sup>, a set of  $2N$ -dimensional nonlinear simultaneous equations in place of Eq. (8) is solved in the main distillation calculation; the liquid flow rates as well as the temperatures at the stages are independent variables of the set of equations. However, it can give better convergence in the simulation to consider the heat balances in an outer iterative loop. Hence, the liquid flow rates are recalculated in the outer loop similar to the nonideality of the solution. The flow chart of calculation is shown in Figs. 1 and 2. As described above, the initial values of temperatures are carefully chosen to obtain the convergence. In the present paper,  $T_1$  and  $T_N$  are carefully chosen, and then an initial set of temperatures is determined by interpolation by using the linear functions of the number of the theoretical stages. The method where three or four temperatures,  $T_1$ ,  $T_{\text{mid}}$ ,  $T_N$ , were given were also tested, however, it did not necessarily give better results.

### 3. Characteristics of Column Having a Feedback Stream

#### 3.1 Column having a feedback stream in isotope separation system in BBI

The column having a feedback stream plays an important role especially in the BBI where a mixture of H-T must be separated into  $H_2$  and  $T_2$ . The molecular specie of HT must not be contained both in the hydrogen stream and in the tritium stream. Kinoshita et al. proposed a two-column cascade for the BBI<sup>(10)</sup>. Figure 3 shows this cascade configuration. The design and operations parameters for the column cascade are presented in Table 1. The feed back stream decomposes the HT supplied into the column to  $H_2$  and  $T_2$  through a equilibrator.

One column having a feed back stream can satisfy the function as the same as that of this two-column cascade. However, the two-column cascade is more excellent than the one-column system, since the tritium inventory is significantly smaller in the two-column system than in the one-column system. As presented in Table 1, the tritium inventory of the two-column cascade is about 8 g. On the other hand, if the one-column system is applied, the tritium inventory is expected to be a few hundred grams<sup>(10)</sup>. In addition, one more disadvantage of the one-column system can be pointed out. In the case where the mixture of H-T is separated by the one-column system, both the top and bottom products are important. Essentially tritium free stream is recovered at the top, while high purity tritium is recovered at the bottom. It is not easy task to maintain the purity of both top and bottom products against disturbances for the feed flow rate and feed composition. For the two-column cascade, the top product stream is more important than the bottom for the column (1) as seen in Table 1. The bottom product stream is more important for the column (2). If the tritium concentration in the top product of the column (2) becomes higher than a designed value, the top stream can be fed to the column (1). Since the flow rate of the top stream of the column (2) is very small in comparison with that of the feed

stream of the column (1) (See Table 1), the column (1) can maintain its separation performance even for this case. From the view point of easiness of the control also, the two-column system is more excellent than the one-column.

Kinoshita et al. reported the characteristics of the column having a feedback stream for the case of separation of H-T. Their conclusions are as follows.

- (i) The side stream location has surprisingly large effects on column performance. If the location is not appropriate, high performance cannot be assured even by an excessively large number of stages and high reflux ratio. The flow rate of the side stream also has large effects.
- (ii) To consider all the factors -viz. (a) the difference in the latent heat of vaporization among the molecular species, (b) the decay heat of tritium, and (c) the nonideality of the solution is found to be important for estimation of accurate purity of product streams.

In the following section, we discuss the effect of flow rate and location of side stream for the column separating H-D system. All the factors described above are considered in the simulation model.

### 3.2 Effect of flow rate of side stream and side stream location

A representative case used for the simulation are listed in Table 2. The first simulation was performed for the case where the column has no feedback stream. Other input specifications are the same as those in Table 2. The composition distribution within the column thus calculated are shown in Fig. 4. As shown in Fig. 4, the concentration of HD has the maximum value at about 70th stage. For this reason, the location of the side stream is determined at 70th stage. Table 2 indicates that the amount of HD supplied into the column is  $\sim 1.0$  mol/h. The second simulation was performed under the conditions: the location of side stream is 70th stage and its flow rate is 1.0 mol/h. Figure 5 shows the composition distribution within the column for this case. The basic pattern of the composition distribution is the same for Figs. 4 and 5. However, for the column having a feedback stream, the concentration of HD steeply decreases from 60th stage, and that of  $D_2$  increases by corresponding values. The third simulation was performed by increasing

the flow rate of side stream from 1.0 to 4.0 mol/h. Figure 6 shows the composition distribution within the column for this case. It is clearly observed that the concentration of HD decreases whole the stages.

Table 3 summarizes the effect of the flow rate of side stream on column performance. The purity of both the top and bottom products improves with increase of the flow rate of side stream. Especially in the range of 0~2 mol/h, significant improvement is observed for the purity of products, while for the range of 2~4 mol/h, increase of product purity becomes appreciably slow. These results are quite understandable if we recall the requirement that the amount of HD decomposed per unit time in the equilibrator be almost equal to the amount of HD fed to the column by the external feed. For the case where the flow rate of side stream is smaller than ~2 mol/h, adequate amount of HD is not decomposed in the equilibrator; so that high purity product is not obtained. Even if a larger flow rate than a sufficient value is applied, the amount of HD that is decomposed at the equilibrator can not exceed the amount of HD fed to the column. In other words, the amount of HD that is decomposed in the equilibrator hardly increases even if the larger flow rate than a sufficient value is applied: in the case where the larger flow rate of side stream is applied, the concentration of HD in the side stream decreases; and the percentage of HD that is decomposed in the equilibrator also decreases. As seen in Figs. 5 and 6, the concentration of HD has the maximum value at ~60th stage for the column with a feedback stream. Then, we carried out a simulation, changing the location of side stream from 70th to 60th. The result is also presented in Table 3. The purity of product is significantly improved.

The effects of flow rate and location of side stream were thus verified for the column separating H-D system. We now consider the determination method for the column parameters such as flow rate and location of side stream. Some calculations revealed that the optimum column parameter can be estimated as follows:

- (i) The flow rate of top and bottom products can be determined from the atom fraction of the external feed stream. If the purity of heavier molecular specie is more important, the bottom flow rate should be smaller than the value determined from the atom fraction.



(ii) Calculate the vapor phase composition distribution within the column which has no feedback stream. The total number of stages and reflux ratio should be determined where the maximum concentration of HD within the column exceeds 99%.

(iii) Find the stage number, which gives the concentration of HD the maximum value.

(iv) Determine the flow rate of side stream from the atom fraction of the external feed stream. As described above, it is expected that the purity of product is hardly improved even if the larger flow rate than a sufficient value is applied. The larger flow rate of the side stream results in a larger flow rate of feed stream. The larger flow rate of the feed stream may deteriorate column performance; since it can disturb the liquid flow within the column. From this point of view also, the flow rate of side stream should be set an appropriate value. At least 20~30% of HD in the side stream is expected to be decomposed into H<sub>2</sub> and D<sub>2</sub> through the equilibrators<sup>(7)</sup>. Hence,

$$\begin{aligned} (\text{Flow rate of the feed back stream}) &= (\text{Flow rate of the external feed stream}) \times \\ &(\text{Concentration of HD}) \times 3\sim 4. \end{aligned} \quad (12)$$

(v) Calculate the vapor phase composition distribution within the column. If the concentration of HD reaches the maximum value at the stage that differs the side stream stage, change the side stream stage to there. Calculate the vapor phase composition distribution within the column again. Examine whether or not the product of purity is improved. The optimum side stream location is a function of the external feed composition. In addition, the HETP is expected to be not necessarily constant along the column height for some cases<sup>(2)</sup>. Hence, in the design stage of the column, it is worthwhile to prepare some different points for the side stream location so that it can be changed.

The characteristics of the column separating H-D system was discussed in this chapter; however, the qualitative aspect of conclusion drawn here can be applied for the column separating H-T or D-T system also.

#### 4. Test Plan (TTA-TP-100-26-R0)

##### 4.1 Purpose

The purposes of this experiment are to:

- (1) Investigate operating conditions similar to those which might be encountered in an ITER-like breeding blanket recovery system. This implies a feed of 99% H<sub>2</sub> and 1% T<sub>2</sub>. In this run D<sub>2</sub> will be used in place of T<sub>2</sub>; results thus obtained will be directly transferable to T<sub>2</sub> operations. Study the influence of varying important column parameters, such as reflux ratio, side stream flow rate, and bottom fraction.
- (2) Investigate transient operation of a single column with recycle equilibration loop designed to effect the reaction:  $2HD=H_2 + D_2$ .
- (3) Obtain steady state composition profiles of one column with a recycle.

##### 4.2 Configuration

The ISS will be used in the new configuration for column I with recycle paths to permit withdrawal of a sample from any tap on column I; equilibration, and reinjection into any other column I tap.

##### 4.3 Subsystem and special materials required

All normal TSTA systems should be available since this is a loop experiment. The RAMAN subsystem is especially required for composition analyses. FCU MSB1/2 must be available for use if necessary. FCU flow paths must be available for recycle of Raman analysis samples. The ISS GC's will not be operated. No special materials are required.

#### 4.4 Personnel required

As this is a loop experiment, all TSTA personnel will be on hand or on call. The facility will be staffed round-the clock. In particular, ISS must be attended any time that changes in column conditions are implemented.

Key ISS personnel for this run are Robert Sherman, David Taylor, Jim Barnes, and Toshihiko Yamanishi.

Key personnel for the Raman subsystem are Robert Sherman, David Taylor, Mary King, and Hirofumi Nakamura.

#### 4.5 Schedule

The schedule is covered by the requirements set forth in the Outline of the test, section 8.0 (below).

#### 4.6 Hazards associated with this test

Since ISS will be used in its new normal configuration, with only Column I kept cold, only the normal hazards associated with ISS are anticipated. These include:

- (i) The column will contain a large quantity of liquid hydrogen/deuterium. Residual quantities of tritium may be present.
- (ii) Liquid nitrogen is used to cool the ISS radiation shield and the helium refrigerator.

#### 4.7 Data requirements

Data will be archived by MDAC and written in the ISS and/or RAMAN log book.

#### 4.8 Outline of the test

#### 4.8.1 Normal ISS operation

The ISS will always be operated within the safety limits defined in the most current revision of the ISS operating procedure, TTA-SOP-103.

The week before this test the following should be accomplished:

- \* Evacuate ISS to 10 Torr; backfill with helium to 800 Torr.
- \* Purge ISS GC lines.
- \* Evacuate ISS to ~2 Torr.
- \* Repeat ISS purge two more times, and then filled with hydrogen gas to 800 Torr.
- \* The ISS and ISS-GAN will be left at 800 Torr of hydrogen.
- \* Evacuate ISS vacuum jacket.
- \* Evacuate ISS refrigerator vacuum jacket.
- \* Evacuate ISS helium transfer line vacuum spaces.
- \* Cool ISS with liq N<sub>2</sub>.
- \* Plug in liq N<sub>2</sub> level controller.
- \* Evacuate ISS-GAN manifold.
- \* Evacuate connecting lines to EXP-2.
- \* Evacuate manifold and Raman cell in Exp-2.
- \* Perform flow meter zero check.
- \* Perform pressure check of ISS pressure gauges.
- \* Start execution of step 5 of ISS Control Program (TTA-TP-121-13).

#### Day 1

- \* Open all helium flow control valves on Column I to ~50%.
- \* All other helium flow control valves to be kept closed.
- \* Start helium refrigerator using two compressors to improve cool down time.
- \* Set helium temperature controller to 23 K.
- \* Evacuate hydrogen in ISS to TWT as Column I is cooling.
- \* Plug in heater controllers for Column I to 117V AC raceway inside ISS control panel; check heater set points.

\* Fill the UTB standard volume by manifolding an H<sub>2</sub> and D<sub>2</sub> cylinder to the D<sub>2</sub> inlet to TP1. With proper purging of the inlet manifold impurities will not be added to Column I. The required Column I fill is 8 moles of H<sub>2</sub> (=179.416 STP liters) and 4 moles of D<sub>2</sub> (=89.708 STP liters). This is equivalent to 612 Torr of H<sub>2</sub> and 306 Torr of D<sub>2</sub> (918 Torr total) in the UTB standard volume which includes an allowance of 10% extra to cover other system volumes.

\* As soon as the standard volume has been filled it may be opened to Column I through EVIA in order to start admitting test gas into the columns.

\* When helium temperature reaches 23 K and column pressures are above 200 Torr, lower refrigerator temperature to 17 K to continue to condense gas from the UTB standard volume.

\* When Column I show signs of liquid in the reboiler,

Close the helium flow valve on the packed section,

Energize reboiler heater to ~2 W.

\* Establish Total Reflux mode by turning on reboiler heaters and setting reboiler power to values listed in Table 4.

\* Change over to Loop Flow mode by opening valves CLIC, CLDA, and CLTB.

\* Establish conditions as given in Table 1. See Note 1 to Table 1.

\* In order to obtain good column profile measurements it is important to maintain the ISS as stable as possible. If column conditions must be changed, small increments should be made. If column pressure is too high or too low, the best adjustment is made by changing the helium refrigerator control temperature, generally in steps of 0.1 K.

\* After stable operation of the loop has been achieved and analyses have been obtained, operating conditions will be changed in one of the following manners (to be determined by the ISS system designer):

(a) Start recycle from tap S1A of Column I through newly installed flow meter/controller and return to tap A2S. Various flows will be tried as listed in Table 5.

(b) Increase or decrease reboiler power to change reflux ratio at appropriate values as listed in Table 5.

(c) Increase or decrease top stream flow rate at appropriate values to obtain bottom flow rate as listed in Table 5.

#### Day 2-4

Adjust Column I conditions as necessary to obtain steady state and observe transient operation. It may be desirable to change the position of the feed, extraction and reinjection taps. Normal extraction will be from tap S1A and reinjection will be into A2S. Normal feed injection will be at feed point A. Operation at various reflux ratios from 2 to 20 may be tried, if time permits.

#### Day 5

Complete data collection and prepare for shutdown.

#### 4.8.2 Shutdown operations

ISS will be isolated from the rest of the loop, and especially from FCU. The contents of Column I will be dumped to the UTB standard volume. Since the column will contain ca. 12 moles of gas, the resultant pressure in the 265 liter volume will be about 918 Torr.

#### 4.8.3 Raman operations

When ISS transients are not being observed, the Raman system will be available to observed transients in the ISS feed(CLIC).

When making a Raman measurement, the column conditions will be recorded manually on the data sheets and in the Raman Log Book. Record at least the following information:

Date and Time

Column Identification

Column Pressure

Differential Pressure

Top Flow

Feed Flow

Reboiler Flow

Reboiler Power

Liquid Level

Table 6 shows Shift assignments for the TSTA Run 93-2.

## 5. Test Results

The experiment was performed by using Column I only among the four columns of the ISS at the TSTA. The temperatures of other columns were kept at  $\sim 77$  K. Table 7 shows specifications of Column I.

### 5.1 Total reflux and recycle operations

#### (i) Day 1

After the Column I was refrigerated to  $\sim 20$  K, a mixture of  $H_2$  and  $D_2$  gases was charged into the column in accordance with the test plan. A mixture gas of  $H_2$  and  $D_2$ , whose amount was about 2 moles, was further added to the column. After the Column I showed signs of liquid in the reboiler, the total reflux operation was initiated. The analysis by the Raman was also initiated.

#### (ii) Day 2

Operation mode was switched from the total reflux to the total recycle. Since the mixture of  $H_2$  and  $D_2$  was charged without through an equilibrator, no HD could be

Date and Time

Column Identification

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Differential Pressure

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#### (ii) Day 2

Operation mode was switched from the total reflux to the total recycle. Since the mixture of  $H_2$  and  $D_2$  was charged without through an equilibrator, no HD could be



observed within the column. In the total recycle mode, a feed stream was supplied through an equilibrator; HD was composed at the equilibrator, so that its inventory within the column gradually increased.

A dynamic simulation of the column during this operation was performed by the computer code. Input specifications of the simulation are shown in Table 8. The experimentally observed values are used in the simulation: the column pressure, flow rate of product streams, initial composition of the gas charged into the column, and liquid holdup in the reboiler. The liquid holdups within the packed section and in the condenser are assumed to be 15% and 2% of their superficial volume<sup>(3)</sup>, respectively. The vapor flow rate at the reboiler can be evaluated from reboiler heater power and latent heat of the hydrogen isotope mixture in the reboiler. Since the simulation model does not consider the heat balance, the reflux ratio can easily be calculated under the condition that the feed stream is supplied as a liquid state. In the simulation, it was assumed that composition within the column at the initial time was uniform. This is because our objective is to obtain qualitative information for the increase of HD inventory within the column. Figure 7 shows dynamic variations of HD and D<sub>2</sub> concentrations in the bottom stream of the column. After initiation of the distillation, the concentration of D<sub>2</sub> in the bottom stream reaches 99.99% within 0.3 hr; and then decreases gradually. Especially during from 20 hours to 40 hours, its concentration relatively and quickly decreases. The concentration of HD increases by corresponding value. In the experiment, appreciable decrease of D<sub>2</sub> concentration in the bottom stream was observed during from 18 hours to 40 hours. Thus, the qualitative tendency for the dynamic behavior of the column could be predicted by the simulation.

## 5.2 Total recycle with a feedback stream operation

After the total recycle operation, the column was put into the operation with a feedback stream. Several runs were carried out by changing the flow rates of side and bottom streams and reboiler heater power in Day 3~5. The composition distribution

within the column was measured with the Raman method. After the experiment was ended, the H-D gas within the column was exhausted to TWT. The shutdown operation was completed in accordance with the test plan.

The dynamic behavior of the column after it was put into the operation with a feedback stream was also simulated by the computer code. Input specification of the simulation is listed in Table 8. In the simulation, the total recycle operation with a feedback stream is initiated from 60 hours after. The simulation result is shown in Fig. 7. After the operation with the feedback stream is initiated, the concentration of HD in the bottom stream quickly decreases; and it reaches a steady state value within 8 hours. In the experiment also, the column needed about 8 hours to reach the steady state. For the column having a feedback stream also, qualitative tendency for the dynamic behavior of the column could be well predicted by the simulation.

An example of composition distribution within the column having a feedback stream was shown in Table 9. The experimental conditions listed in Table 9 are almost equal to those in Table 2, if it is assumed that the HETP value is 5 cm and is constant along the column height<sup>(4)</sup>. The calculated results were already shown in Table 3. It is observed that the concentration of H<sub>2</sub> in top stream and that of D<sub>2</sub> in bottom stream experimentally observed are in rough agreement with the calculated results. The column having the feedback stream gives us high purity D<sub>2</sub>. In contrast with this experimental result, the column without the feedback stream showed that the major component in the bottom stream was HD. The effect of the feedback stream on the column performance was thus verified from the experimental results.

## 6. Conclusion

The characteristics of the column having a feedback stream were discussed by the simulation works. Several computer codes were arranged; slight modification was made on these codes to obtain good convergence in the simulation and to consider the several factors such as the dead volumes and the by-pass flows in the column.

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The flow rate of side stream affects the column performance; a larger flow rate than a sufficient value does not give good column performance. Since it is expected that the larger flow rate of feed stream deteriorates the column performance, the flow rate of feedback stream must be set an appropriate value. The location of side stream has the significant effect on the column performance. To choose the optimum side stream location is more important than the increases of flow rate of side stream, total number of theoretical stages, and reflux ratio for improvement of the column performance. A method for determination of design and operational parameters of the column was also developed: (1) location and flow rate of side stream, and (2) top and bottom flow rates.

The experiment for the column having a feedback stream was successfully performed with the H-D system at the TSTA. In the case where the column was operated with the feedback stream, high purity  $D_2$  whose concentration was in agreement with calculated result was obtained from the bottom stream. Thus, experimental results with the H-D system verified the effect of feedback stream on the column performance. In further experiments, it is desired to examine the effect of feedback stream in detail; the effect of flow rate and location of side stream should be systematically examined.

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## NOMENCLATURE

$F$  = flow rate of feed stream (mol/h)

$H_L$  = Liquid holdup at stage (mol)

$K$  = vapor-liquid equilibrium constant (-)

$L$  = liquid flow rate (mol/h)

$N$  = total number of theoretical stages (-)

$T$  = temperature at stage (K)

$t$  = time

$V$  = vapor flow rate (mol/h)

$x$  = mole fraction in liquid phase (-)

$y$  = mole fraction in vapor phase (-)

$z$  = mole fraction in feed stream (-)

$Z$  = atom fraction in side stream (-)

$\alpha$  = setting value (The change of temperature should be smaller than this value in iteration by the Newton-Raphson method.) (K)

## Subscript

$D$  = deuterium

$H$  = hydrogen

$i$  = component

$j$  = stage

mid = middle of column

## Superscript

ex = external feed

in = internal feed

Table 1 Design and operation parameters for column cascade in BBI

	Column (1)	Column (2)
Number of total theoretical stages (-)	40	70
Feed stage number (-)	25	40
Side stream stage number (-)		45
Top flow rate (mol/h)	98	1.5
Bottom flow rate (mol/h)	2	0.5
Side stream flow rate (mol/h)		4
Reflux ratio	2.5	7
Diameter of column (cm)	5.0	1.0
Height of column (m)	1.9	3.4
Inventory of tritium (g)	3.87	4.35
Top composition	H <sub>2</sub> : 1.000 HD : 3.56x10 <sup>-6</sup> HT : 8.47x10 <sup>-7</sup>	H <sub>2</sub> : 0.981 HD : 1.85x10 <sup>-2</sup> HT : 5.20x10 <sup>-4</sup>
Bottom composition	H <sub>2</sub> : 0.487 HD : 1.47x10 <sup>-2</sup> HT : 0.496 D <sub>2</sub> : 1.39x10 <sup>-6</sup> DT : 1.02x10 <sup>-4</sup> T <sub>2</sub> : 1.98x10 <sup>-3</sup>	H <sub>2</sub> : 0.0 HD : 0.0 HT : 3.06x10 <sup>-6</sup> D <sub>2</sub> : 2.42x10 <sup>-7</sup> DT : 2.03x10 <sup>-3</sup> T <sub>2</sub> : 0.998

Table 2 Input specifications of steady state simulation  
for the column having a feedback stream

Pressure (Torr)	760
Flow rate (mol/h)	
Top	7.5
Bottom	0.18
Feed	7.68
Side stream	4.0
Number of total theoretical stages (-)	80
External feed stage number (-)	50
Internal feed stage number (-)	50
Side stream stage number (-)	70
External feed composition (-)	
H <sub>2</sub>	0.878
HD	0.117
HT	$1.0 \times 10^{-7}$
D <sub>2</sub>	$4.82 \times 10^{-3}$
DT	$1.0 \times 10^{-5}$
Reflux ratio (-)	10.0

Table 3 Effect of flow rate and location of side stream on column performance

Flow rate of side stream (mol/h)	0.0	0.5	1.0	2.0	3.0	4.0	2.0*
Mole fraction of H <sub>2</sub> in top product (-)	0.8992	0.9135	0.9164	0.9173	0.9175	0.9177	0.9182
Mole fraction of D <sub>2</sub> in bottom product (-)	0.2057	0.8027	0.9218	0.9583	0.9693	0.9747	0.9954

\* The side stream is withdrawn from 60th stage.



Table 4 Initial ISS conditions for Column I

Pressure (Torr)	760
Top Flow (cm <sup>3</sup> /min)	2940
Feed Flow (cm <sup>3</sup> /min)	3000
Bottom Flow (cm <sup>3</sup> /min)	60
Reboiler Power (W)	~20
H <sub>2</sub> : D <sub>2</sub> feed ratio	99:1
Reflux ratio	~5

\* These conditions are approximate and final values can only be determined after Raman analysis of the actual ISS/CLIC feed stream.

\* Front panel Flow meters readout is in either liters/min or cm<sup>3</sup>/min depending on the range: MDAC display is in cm<sup>3</sup>/min.

Table 5 Probable parameter variations

Parameter	Range	Note
Side stream flow	0, 0.02, 0.04, 0.1, 0.2, 0.5, 1.0	Fraction of main feed
Reboiler flow	0.008, 0.01	Fraction of main feed
Reflux ratio	2, 5, 10, 20	

Table 6 Shift assignments for the TSTA Run 93-2

April 12-16, 1993

	Shift A 0730-1530	Shift B 1530-2330	Shift C 2330-0730
Test Dir	Willms	Bartlit	Loop Operator
Alt TD	Nasise	Barnes	
Loop Operator	Wilhelm	King	Harbin
Facility op.	Dahlin	Jenkins	Hamerdinger
TSTA Subsystem			
ISS	Sherman/Yamanishi /Suzuki	Barnes/Hayashi	Nakamura
FCU	Willms	(Willms)	
UTB	Nasise	Jenkins	
TPU	Dahlin	Jenkins	
LIO	Nasise	Jenkins	
TWT	Dahlin/Wilhelm	(Nasise)	
ETC	Harbin	King	
SEC		Barnes	
IMS	Nasise	Willms	
MDAC	Cole	(Cole)	
GAN-RAMAN	Sherman/Taylor/ Yamanishi	Hayashi	Nakamura

\* All others will be on the A shift and subject to call as needed.

\* Some personnel on Shifts A&B may shift hours between shifts (with the cognizance of the TD) depending on specific activities under way.

Table 7 Specifications of Column I

Inner diameter (cm)	2.84
Packed height (cm)	411
Packing material	Heli-Pak (4.4x4.4x2.3 mm)
Superficial volume (cm <sup>3</sup> )	
Packed section	2494
Condenser	1915
Reboiler	499
Total	4908
Sampling location	Bottom
(The column has 7 sampling points.)	15" from bottom
	39" from bottom
	55" from bottom
	89" from bottom
	129" from bottom
	Top

Table 8 Input specifications of dynamic simulation

Pressure (Torr)	760
Flow rate (mol/h)	
Top	7.5
Bottom	0.18
Feed	7.68
Side stream	4.0
Number of total theoretical stages (-)	80
External feed stage number (-)	50
Internal feed stage number (-)	50
Side stream stage number (-)	70
Holdup (mol)	
Condenser	1.32
Packed section	0.18
Reboiler	0.7
Reflux ratio (-)	7.5
Initial composition of gas charged into the column	H:D = 2:1

Table 9 Example of experimental observation for the column  
having a feedback stream

Pressure (Torr)	765		
Flow rate (mol/h)			
Top	7.5		
Bottom	0.18		
Feed	7.68		
Side stream	4.0		
External feed location	55" from bottom		
Internal feed location	55" from bottom		
Side stream location	15" from bottom		
Power of reboiler heater (W)	20		
Composition distribution			
Sampling location	H <sub>2</sub>	HD	D <sub>2</sub>
Top	0.943	0.035	0.022
129" from bottom	0.956	0.031	0.013
89" from bottom	0.167	0.819	0.014
55" from bottom*	0.441	0.306	0.253
39" from bottom	0.054	0.784	0.162
15" from bottom	0.004	0.585	0.402
Bottom	0.007	0.025	0.936

\* This experimental observations were expected to show the composition of the combined feed stream that consisted of the external and internal feed streams.

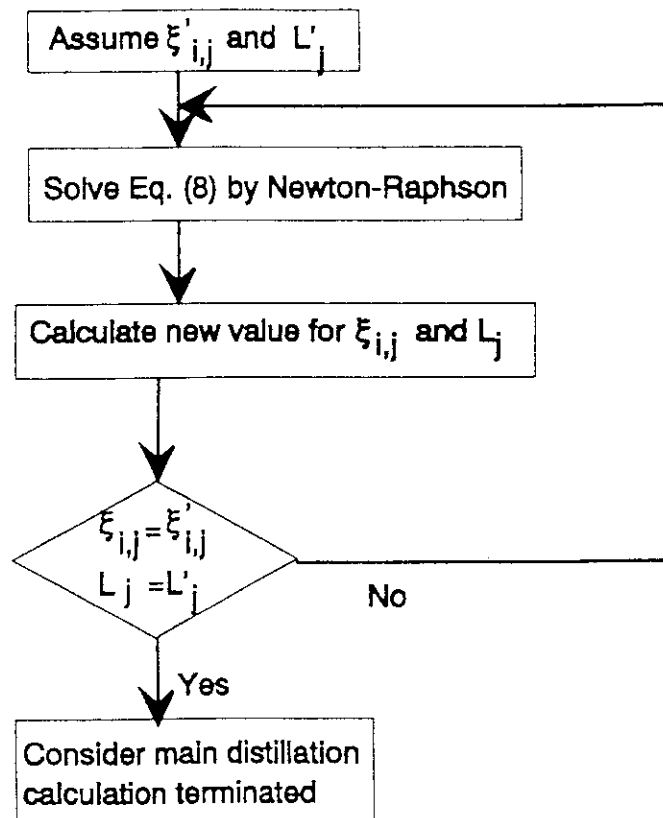
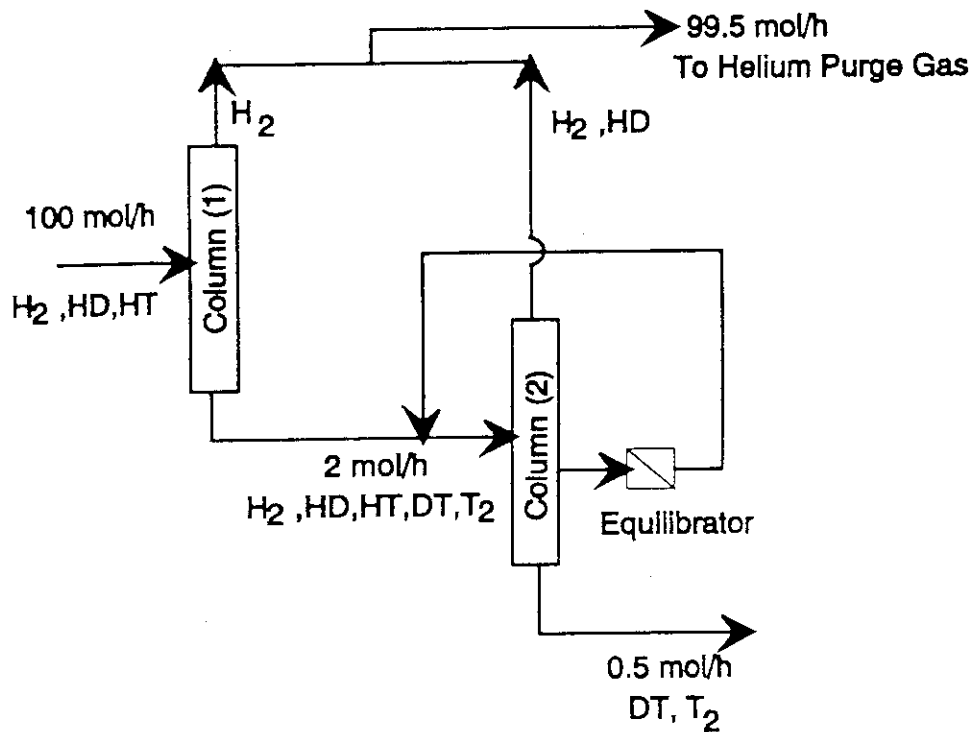


Fig. 1 Flow chart for main distillation calculation.

Fig. 3 Two-column cascade developed for the BBI by Kinoshita et al.<sup>(10)</sup>.

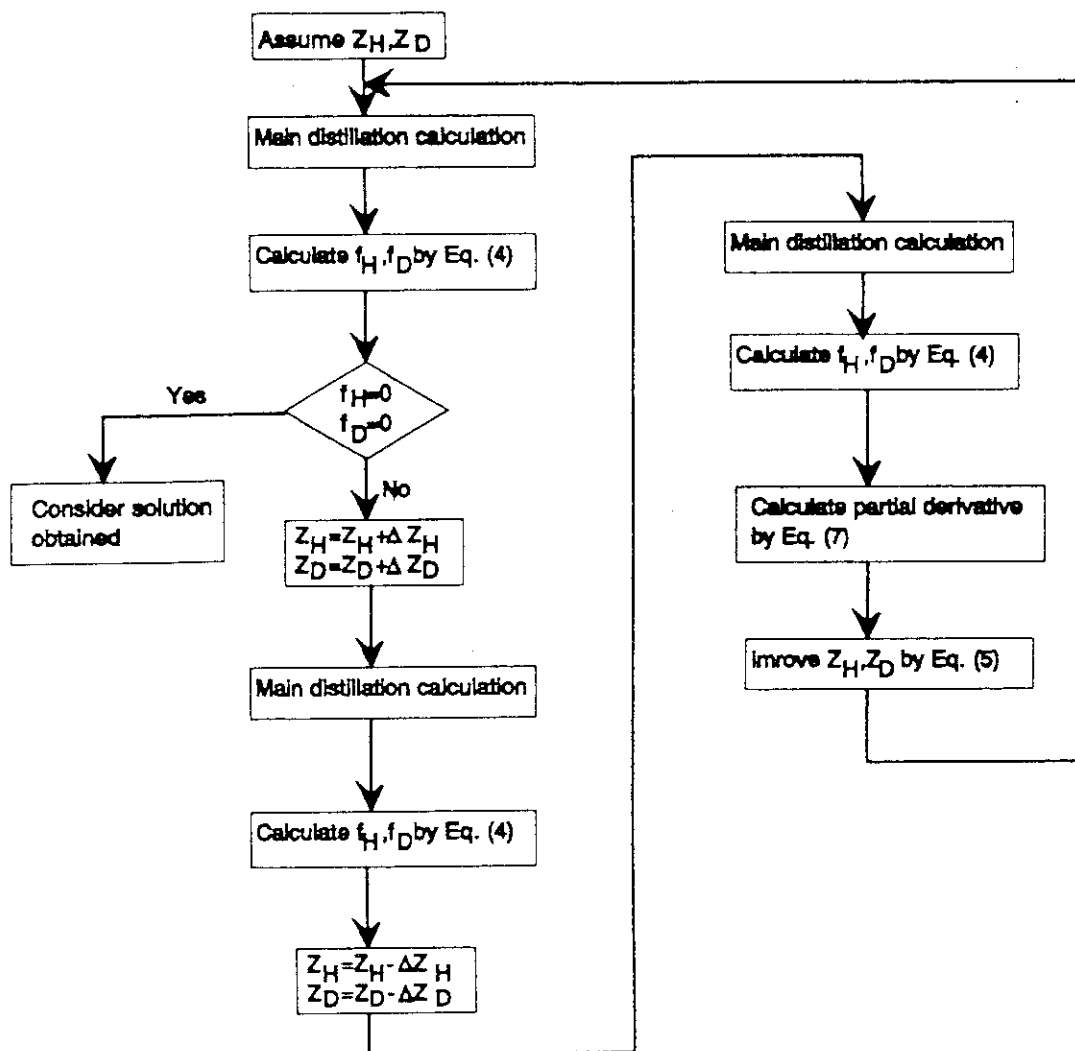


Fig. 2 Flow chart for whole calculation.

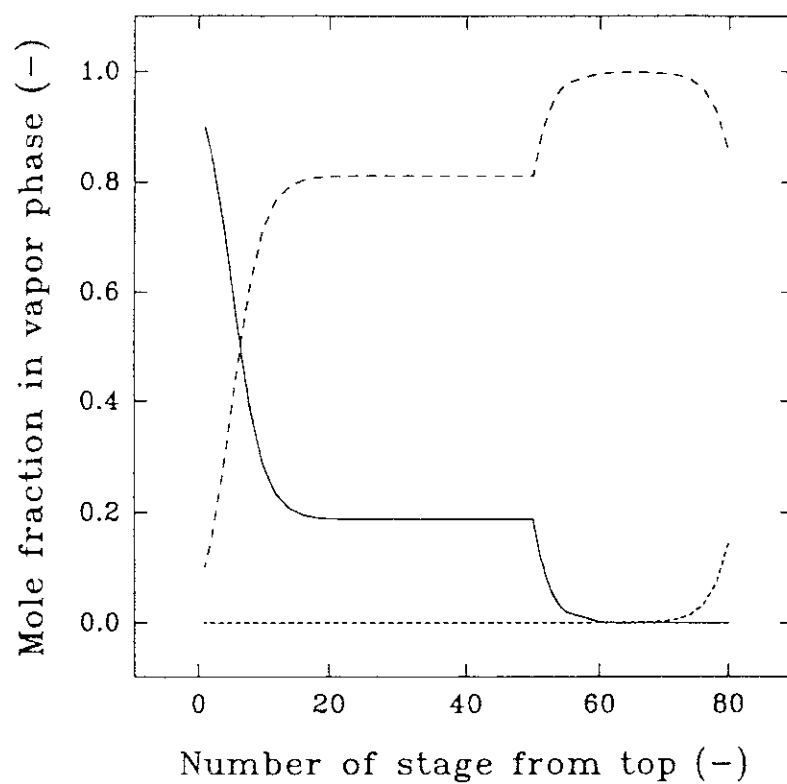


Fig. 4 Composition distribution within the column without feedback stream:  
 —,  $H_2$ ; ---,  $HD$ ; ...,  $D_2$ .

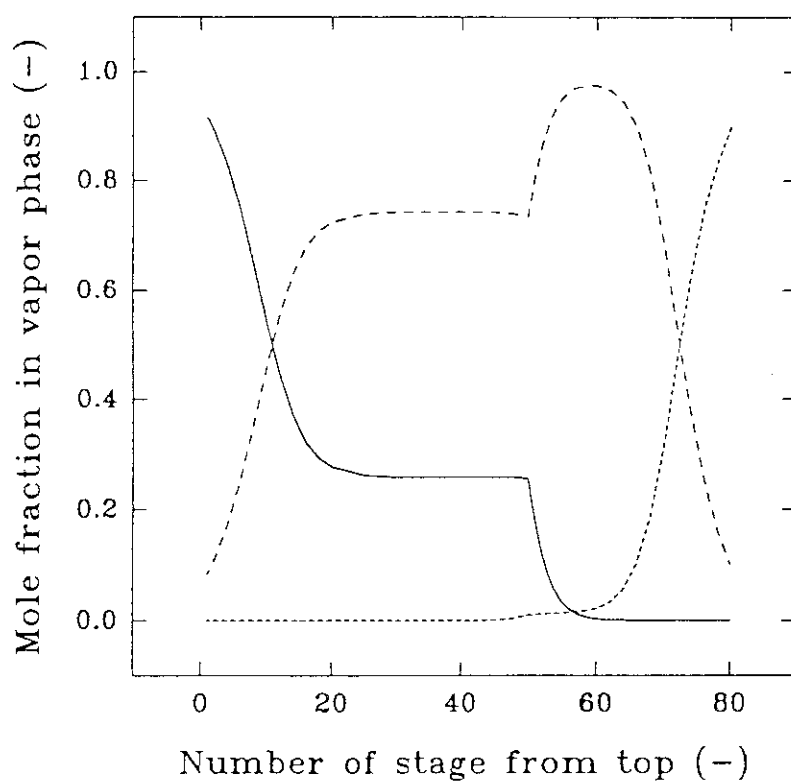


Fig. 5 Composition distribution within the column in the case where flow rate of side stream is 1.0 mol/h:  
 —,  $H_2$ ; ---,  $HD$ ; ...,  $D_2$ .

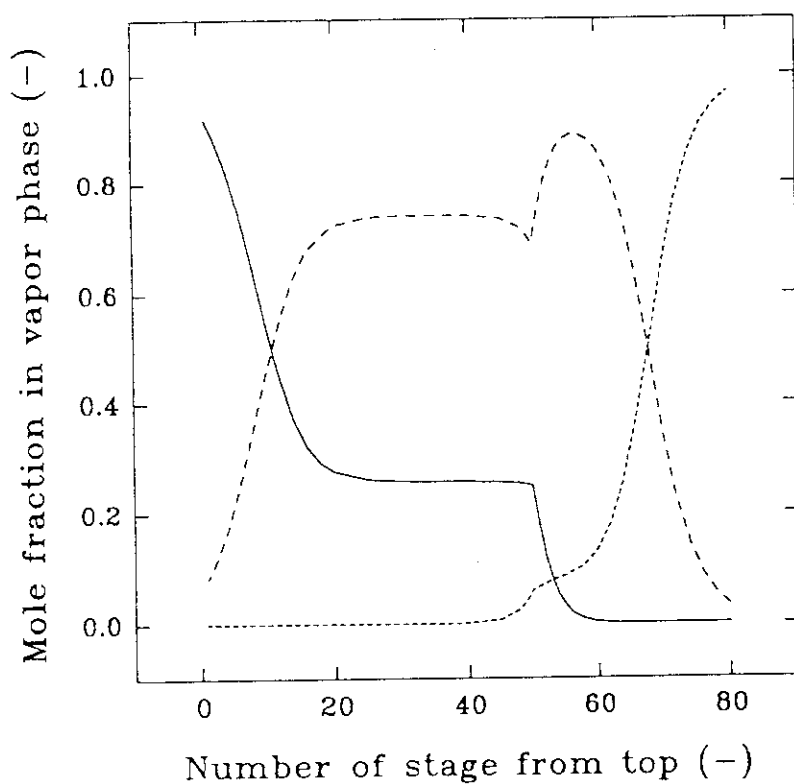


Fig. 6 Composition distribution within the column where flow rate side stream is increased from 1.0 to 4.0 mol/h: —,  $H_2$ ; ---, HD; ...,  $D_2$ .

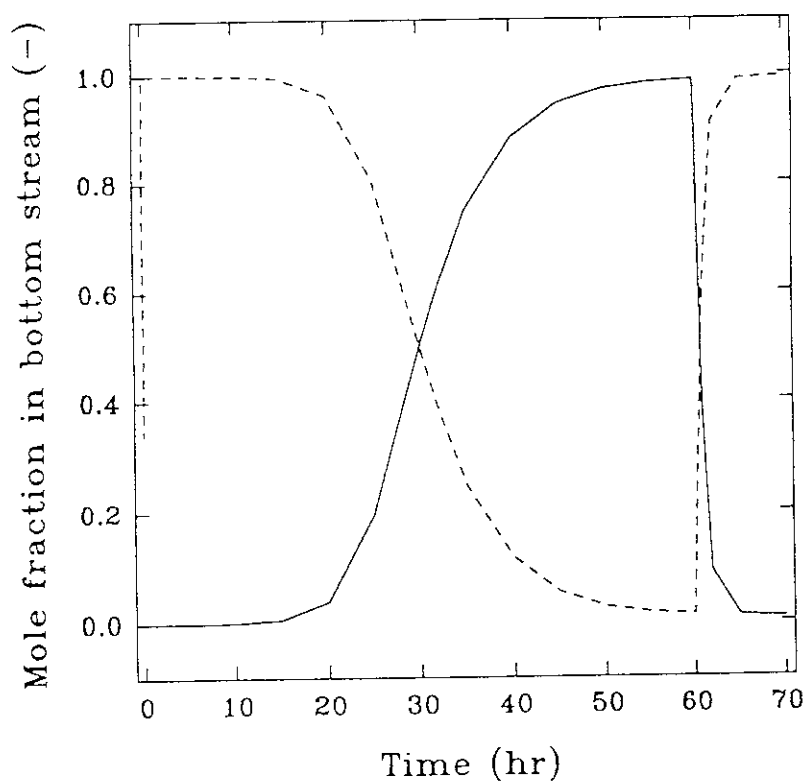


Fig. 7 Dynamic variation of composition in bottom stream: —, HD; ---,  $D_2$ .