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DESIGN CONCEPT OF A CRYOGENIC
DISTILLATION COLUMN CASCADE
FOR A ITER SCALE FUSION REACTOR

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Toshihiko YAMANISHI, Mikio ENOEDA and Kenji OKUNO

日本原子力研究所
Japan Atomic Energy Research Institute

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Design Concept of a Cryogenic Distillation Column Cascade
for a ITER Scale Fusion Reactor

Toshihiko YAMANISHI, Mikio ENOEDA and Kenji OKUNO

Department of Fusion Engineering Research

Naka Fusion Research Establishment

Japan Atomic Energy Research Institute

Naka-machi, Naka-gun, Ibaraki-ken

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A column cascade has been proposed for the fuel cycle of a ITER scale fusion reactor. The proposed cascade consists of three columns and has significant features: either top or bottom product is prior to the other for each column: it is avoided to withdraw side streams as products or feeds of down stream columns: and there is no recycle steam between the columns. In addition, the product purity of the cascade can be maintained against the changes of flow rates and compositions of feed streams just by adjusting the top and bottom flow rates. The control system has been designed for each column in the cascade. A key component in the prior product stream was selected, and the analysis method of this key component was proposed. The designed control system never brings instability as long as the concentration of the key component is measured with negligible time lag. The time lag for the measurement considerably affects the stability of the control system. A significant conclusion by the simulation in this work is that permissible time for the measurement is about 0.5 hour to obtain stable control. Hence, the analysis system using the gas chromatography is valid for control of the columns.

Keywords: ITER, Fuel Cycle, Cryogenic Distillation, Column Cascade, Control System,
Hydrogen Isotope Separation, Tritium, Gas chromatography, Feed Back

I T E R規模の核融合炉の深冷蒸留塔カスケードの概念設計

日本原子力研究所那珂研究所核融合工学部

山西 敏彦・榎枝 幹男・奥野 健二

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I T E R規模の核融合炉における燃料サイクルのための深冷蒸留塔カスケードを提案した。カスケードは3本の塔からなり、以下の重要な特長を持っている。カスケードを構成するすべての塔に対し、塔底あるいは塔頂プロダクトどちらかがより重要である。サイドカット流を、プロダクトあるいは下流の塔のフィード流としていない。塔間のリサイクル流がない。加えてカスケードのプロダクト純度は、フィード流の組成及び流量変化に対して、塔頂及び塔底流量を調整するだけで一定に保つことができる。カスケードを構成する各々の塔に対して、制御系を設計した。重要なプロダクト流中の鍵となる成分を選択し、その成分の分析手段を提案した。設計した制御系は、鍵となる成分の濃度が遅れ時間なく測定できる限り不安定とはならない。分析に要する遅れ時間は、制御系の安定性に大きな影響を与える。一つの重要な解析による結論として、制御系を安定させるために許される分析時間が、約30分であることが判明した。ガスクロマトグラフにより、水素同位体の分析を30分間で行うことは可能であり、制御系の分析手段として用いることは可能である。

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1. Introduction

In the fuel circulation system of a fusion reactor, hydrogen (H) must be removed from the deuterium and tritium mixture, and high purity stream of deuterium (D) is required. Cryogenic distillation is recognized to be best suited to the needs in this fuel circulation system. Kinoshita et al. [3,4] have proposed several cascades for the main fuel circulation and bleeder blanket systems. They discussed effect of disturbance such as variation of compositions and flow rates of feed streams on the performance of these cascades. Next subject to be studied is design of a column cascade which process all the hydrogen isotope mixtures from the torus exhaust, neutral beam injector (NBI), and bleeder blanket. The pellet injector (PI) propellant gas also, which is expected to contain small amount of tritium, may be processed by the column cascade. Although some column cascades have already been proposed for this purpose [5,6], no detailed information for simulation was described in the reports: whether or not some factors such as nonideality of hydrogen isotope solution and decay heat of tritium were considered. Another serious problem is that no concern is paid to control method of product purity in the design of column cascade. The purity of product streams in the fuel cycle system is considerably severe [5,7], so that control method must be carefully considered. For instance, the cascade should be designed where either top or bottom product stream is important for each column, since it is difficult to maintain purity of both top and bottom products.

For distillation columns used in the hydrocarbon processing, purity of product stream can be expected from temperature at top or bottom of the column [8]. However, for the cryogenic distillation columns, it is difficult to know the purity of product from the temperature. Exact measurement of the temperature is not easy task at a cryogenic temperature range (~ 20 K). In addition, the temperature of the column hardly varies even if the purity of product deteriorates to some extent. For instance, tritium concentration in a hydrogen waste gas should be maintained at extremely low values (Atom fraction of T should be less than $\sim 1 \times 10^{-7}$.) [7]. If the tritium

concentration in the waste gas increases to $\sim 1 \times 10^{-5}$, it would cause a serious problem from the view point of tritium release; however, the temperature hardly varies. It is also a significant subject for the design of control system what analysis method should be applied.

The present paper proposes a configuration of column cascade for the fuel cycle of a ITER scale fusion reactor. In recent design of ITER, NBI is not applied. However, there is possibility that NBI is applied for a future fusion reactor. Hence, we design a column cascade that processes the gas from NBI and produces high purity D_2 . A significant purpose of the present study is to consider control method of the cascade. A key component whose concentration should be controlled differs in each column. An appropriate analysis method and the key component should be selected for each column. Stability of the control system is also discussed for a representative case.

2. Column cascade proposed

2.1 Basic philosophy for design of column cascade

In the fuel circulation system of the fusion reactor, requirement for purity of product is very strict [5,7] in comparison with ordinary columns used in the hydrocarbon processing. Hence, we must pay attention to the following discussions.

- (i) The cascade should be designed where either top or bottom product of a column is important, since it is difficult to maintain purity of both top and bottom products at very high values.
- (ii) It is not desirable to return products of down stream columns to upper stream columns. For this case, a change of operational conditions of the down stream column such as the flow rate of top or bottom stream affects the performance of the upper

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2. Column cascade proposed

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- (i) The cascade should be designed where either top or bottom product of a column is important, since it is difficult to maintain purity of both top and bottom products at very high values.
- (ii) It is not desirable to return products of down stream columns to upper stream columns. For this case, a change of operational conditions of the down stream column such as the flow rate of top or bottom stream affects the performance of the upper

stream column. The change of performance of the upper stream column affects that of the downstream columns again. The system is hardly stabilized because of the infinity loop.

(iii) It should be avoided to withdraw a side stream as a product or a feed of a downstream column. In the design of cryogenic distillation columns, optimum stage numbers of feed and side streams are obtained by simulation. The locations of feed and side streams of actual columns would be determined under the assumption that the HETP value for each theoretical stage is constant. However, it was observed that the HETP value for each theoretical stage was subject to change with operational conditions of the columns as reported in our previous papers [9,10]. This means that the stage numbers of feed and side streams of actual columns are subject to change with operational conditions. Even if the feed stage number varies to some extent, the purity of top and bottom products can be maintained at design values by adjusting the flow rates of product streams and reflux ratio. However, if the side stream is withdrawn as a product, the change of its stage number would cause a large variation of product purity. In addition, it is difficult to control the purity of the side stream in comparison with that of top or bottom stream. For the case where the side stream is withdrawn as a feed stream of a downstream column, the change of stage number of side stream results in a large variation of feed composition. To maintain the purity of products of the downstream column is not an easy task for this case.

The column cascades previously reported by several workers [5,6] for the fuel cycle did not satisfy these requirements. For an example, the cascade proposed by Sze et al [5] is shown in Fig. 1. There are many recycle streams between columns. The side streams are withdrawn as a product and a feed stream. The cascade does not satisfy the requirements of (ii) and (iii) described above. The four-column cascade at the Tritium Systems Test Assembly (TSTA) of Los Alamos National Laboratory is a promising system that satisfies the requirements described above. However, as Kinoshita has pointed out [3], the cascade can accept only a small amount of H ($\sim 1\%$

in feed streams). This limitation for the concentration of H in the feed stream is a serious disadvantage of the TSTA cascade.

2.2 Determination of cascade configuration

The column cascade proposed is illustrated in Fig. 2. It consists of three columns and an equilibrator. All the requirements described in the preceding section are satisfied in this column cascade. Design values for flow rates of hydrogen isotope gases reported by ITER-EDA and CDA are summarized in Table 1[5]. The flow rate of the gas from the blanket has not been determined in ITER-EDA. Hence, we use the value by ITER-CDA. Deuterium contained in natural hydrogen is taken into account on composition of the gas from the blanket. In the recent design of ITER, NBI is not applied. However, there is possibility that NBI is used for a future fusion reactor. In this case, a part of the gas from NBI would be sent to the column cascade, and the residual gas would directly be recycled to NBI. We assume that the flow rate of the gas sent from NBI is 100 mol/h. The values by ITER-CDA are used as the concentration of this gas. An amount of gas for PI is ~400 mol/h in ITER-CDA. If it is required to process a part of PI gas, the flow rate of the gas sent to the column cascade may be ~100 mol/h similar to the case of NBI. It is estimated in ITER-EDA that a large amount of hydrogen gas (~500 mol/h) containing tritium is sent to the column cascade from water processing systems. The tritium concentration of this hydrogen gas is much lower than that of PI gases. Hence, if the column cascade can accept the gas from PI whose flow rate is 200~300 mol/h, the column cascade can easily accept the gas from the water processing systems (~500 mol/h). For these reasons, we assume that the flow rate of the gas from PI is 300 mol/h. The flow rate of the plasma exhaust gas is estimated to be 160 mol/h in ITER-EDA. It is reported that only 10% of the plasma exhaust gas (16 mol/h) is processed by the column cascade from the viewpoint of maintaining hydrogen concentration in a D-T fuel at a

desirable value. If the combustion rate of the D-T fuel in a plasma is increased, a large amount of the plasma exhaust gas should be processed by the column cascade. We select the value of ITER-CDA (71.4 mol/h) as a representative flow rate. The flow rates and compositions of the gases sent from NBI, PI, and the plasma were thus determined as shown in Fig.2. Although there is uncertainty for the compositions and flow rates of these streams, the column cascade can produce required purity of products by adjusting the some design parameters : flow rates of top, bottom, and side streams; reflux ratio; and inner diameter of the columns.

The hydrogen isotope gases from the bleeder blanket and PI are charged into column (1). The top product of column (1), which is hydrogen containing small amount of HD, is recycled to the helium purge gas and pellet injector. The three streams, the bottom product of column (1), hydrogen isotope gases from torus exhaust and NBI, are fed to column (2). The essentially tritium free stream is exhausted from the top of column (2). The atom fraction of T in the top stream of column (2) is $\sim 9 \times 10^{-8}$. High purity deuterium is obtained from top of column (3). The purity of D is 99.96%, and atom fraction of T in the bottom stream of column (3) is ~ 0.61 .

The performance of each column was simulated by a computer code to determine the design specifications of the columns. The computer code considers all the factors which are required for the simulation of behavior of the columns: (i) nonideality of hydrogen isotope solutions; (ii) differences in latent heat of vaporization among the isotopic species; and (iii) decay heat of tritium. The basic equations and calculation procedure of the code are described in Appendix. Design and operational parameters of the columns thus determined are shown in Table 2. The resultant top and bottom compositions of each column are also given in Table 2. The inner diameter, height, and tritium inventory of the column are estimated under the following assumptions: (i) the HETP value is 5 cm [11]; (ii) the vapor velocity within the column is ~ 15 cm [4]; and (iii) the liquid holdup in the packed section is $\sim 10\%$ of its superficial volume [12]. Larger packings are used for a column whose inner diameter is larger. Additionally, the vapor velocity would be set at a higher value. The HETP value could

be larger, and the liquid holdup could be smaller than 10%. However, the constant values are used for estimating the tritium inventory of the column because of lack of experimental data. The liquid holdup in the reboiler is assumed to be five times larger than that for a theoretical stage in the packed section [3,4]. The liquid holdup in the condenser is assumed to be equal to that for a theoretical stage [3,4].

2.3 Separation performance of the cascade

As discussed in the preceding section, in the fuel cycle of fusion reactor, there is uncertainty for the flow rates and compositions of feed streams. In addition, the HETP value for each theoretical stage can possibly vary: feed and side stream stage numbers are subject to change [9]. It is important to examine the performance of cascade for these disturbances.

The column (1) can easily be controlled by adjusting the top and bottom flow rates even if the flow rates and compositions of feed streams (from bleeder blanket and PI) appreciably vary. The column (3) also can produce the high purity D by adjusting some operational parameters against the change of composition and flow rate of feed stream. The column (2) is operated with a feedback stream. Kinoshita et al. [13] have reported that location of side stream has a large effect on performance for the column separating H and T with a feedback stream. For these reasons, the effect of location of feed and side stream on the performance was first examined for the column (2). Simulations were carried out by changing the numbers of side and feed streams by ± 10 . The resultant atom fractions of T and H in top and bottom streams are shown in Table 3. The atom fraction of T in the top and mole fraction of HT in the bottom can be maintained at sufficient low values just by adjusting the flow rates of top and bottom even if the stage numbers of side and feed streams vary by ± 10 . For the column (2), the performance is relatively insensitive for the location of side stream. The effect of flow rate and composition of feed stream on the performance of column (2) was then

examined for two cases: the flow rate from NBI decreases from 100 to 50 mol/h; and the concentration of H in the feed stream from torus exhaust increases from 1 to 5%. As seen in Table 3, the high performance of column (2) can be maintained just by adjusting the top and bottom flow rates for these two cases also. It is thus proved that the column (2) has great flexibility against the variations of feed streams (flow rates and compositions) and the HETP value for each theoretical stage.

The tritium inventory is one of the key subjects for design of column cascade; it greatly depends on the product purity required. In the proposed cascade, tritium mainly exists in the column (3). Simulations were carried out for three representative cases: (case 1), it is acceptable that the atom fraction of T in the D-T stream is ~ 0.5 ; (case 2), an enriched tritium stream ($>80\%$) is required as a product; and (case 3), the atom fraction of T in the deuterium stream must be decreased to a lower value. The simulation results are shown in Table 4. For case 1, a smaller reflux ratio can be applied for the column (3), and the tritium inventory within the column decreases to $\sim 1/3$. As a solution of case 2, high purity D and T can simultaneously be obtained by the operation of column (3) with a feedback stream. The purity of D is 99.96% and that of T is 90% as indicated in Table 4. However, a larger inner diameter and height are required, and the column has 2 times larger tritium inventory. The atom fraction of T in the top stream of column (3) can be decreased to $\sim 5 \times 10^{-5}$ by increasing the number of total theoretical stages or reflux ratio. The concentration of HT in the top stream of column (3) can not be decreased by increasing the number of total theoretical stage or reflux ratio, so that it is impossible to decrease the atom fraction of T to a value less than $\sim 5 \times 10^{-5}$. To satisfy the requirement of case 3, it is practical to add a column at the down stream of the column (3): The top stream of column (3) is fed to column (4) through a equilibrator. The column (3) is operated with a feedback stream for the case 2. The composition of top stream of column (3) for the case 2 is almost equal to that without the feedback stream as seen in Tables 2 and 4. A column whose design specifications are almost identical to the column (4) should be added at the down stream of column (3) with a equilibrator for the case where both the

requirements of cases 2 and 3 must be satisfied also. The atom fraction of T in the D stream can be decreased to $\sim 4 \times 10^{-8}$ by adding the column (4). In addition, the tritium inventory of the whole system hardly increases by adding the column (4). However, the increase of number of columns causes difficulty for control of the whole system. These simulation results thus give us a significant conclusion that the product purity should be carefully determined: the atom fraction of T in the D-T stream is chosen as low as possible (The value of ~ 0.5 is desirable from the viewpoint of reduction of tritium inventory.); the atom fraction of T in the D stream is chosen as large as possible.

3. Design concept of control system for column cascade

3.1 Basic design of control system

The column parameters which must be controlled are the purity of product, liquid level of the reboiler, and pressure. Both the temperature and flow rate of refrigerant helium gas can control the column pressure; the temperature of refrigerant helium gas more strongly affects the column pressure. For this reason, the temperature of refrigerant gas is applied as the manipulated variable for the control of column pressure.

Figure 3 shows a typical control system in the case where the top product is prior to the other. The column pressure is controlled by adjusting the heater power installed at the refrigerant helium gas: temperature of the refrigerant helium gas. The flow rate of top stream is selected as the manipulated valuable for the control of top product, [14]. Although the vapor velocity within the column (reflux ratio) can also be a manipulated valuable for the control of product purity, it has an upper limit; since a

requirements of cases 2 and 3 must be satisfied also. The atom fraction of T in the D stream can be decreased to $\sim 4 \times 10^{-8}$ by adding the column (4). In addition, the tritium inventory of the whole system hardly increases by adding the column (4). However, the increase of number of columns causes difficulty for control of the whole system. These simulation results thus give us a significant conclusion that the product purity should be carefully determined: the atom fraction of T in the D-T stream is chosen as low as possible (The value of ~ 0.5 is desirable from the viewpoint of reduction of tritium inventory.); the atom fraction of T in the D stream is chosen as large as possible.

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larger vapor velocity would cause the flooding. The flow rate of the top stream is more excellent manipulated valuable for the control of top product. The liquid level in the reboiler is controlled by the heater power at the reboiler and the flow rate of bottom stream. Increase of the heater power at the reboiler means that of the vapor velocity within the column. The liquid holdup within the packed section increases with the vapor velocity, so that the liquid level in the reboiler decreases[12]. As described above, the vapor velocity has an upper limit. Hence, in the case where the liquid level in the reboiler still increases in spite of adjustment of the vapor velocity; the flow rate of the bottom stream is increased. We call this control mode here in after Mode 1. In the case where the purity of bottom product is prior, the bottom flow rate should be chosen as the manipulated valuable for the control of product purity in place of the top flow rate. The liquid level in the reboiler is adjusted by the heater power at the reboiler and flow rate of top steam similar to the control concept of the Mode 1.

The column having a feed back stream plays an important role of the proposed cascade. The molecular specie of HT must not be contained both in the top and bottom streams. The feed back stream decomposes the HT supplied into the column to HD and DT through a equilibrator ($HT + D_2 \rightarrow HD + DT$). Figure 4 shows a control system of this column having a feed back stream. The top product purity is considered to be prior to the other. The pressure, liquid level in the reboiler, and purity of top product are controlled by the same concept of the Mode 1. A major concern should be maid how do we choose the flow rate of the feed back stream. The feed back stream has a function of decomposition of HT. Its flow rate depends on the amount of HT supplied to the column. Hence, the flow rate of the feed back stream should be controlled by analysis results for concentrations of HT in the external feed and side streams. The flow rate of the feed back stream can be determined from the following equation:

[Flow rate of the feed back stream] \equiv [Concentration of HT] / {[concentration of HT in the side stream before thorough the equilibrator]-[concentration of HT in the side stream after through the equilibrator]} \times [Flow rate of the external feed stream] . (1)

As long as the flow rate of the feed back stream is set an appropriate value for the amount of HT supplied into the column, we can regard the column having the feed back stream as the ordinary column. Hence, the Mode 1 can be applied for controlling other valuables. We call this control mode here in after Mode 2.

3.2 Control system of each column in the cascade

3.2.1 Column (1)

The most critical task for Column (1) is to keep the tritium concentration in the top product within an adequately small range. Hence, the Mode 1 is applied for this column. A key component is considered to be HT in the top product.

As analysis instruments of hydrogen isotopes, we can mention ion chamber, gas chromatograph, and raman. The raman method is attractive, since it requires only a few minutes for measurement of a sample, and does not make waste gas. However, as seen in Table 3, the concentration of HT in the top stream is extremely lower than limitation of analysis of the raman [14]. In addition, concentrations of DT and T₂ are negligible in comparison with that of HT. The ion chamber is sufficient and appropriate as the analysis instrument.

3.2.2 Column (2)

The Mode 2 is applied for the column (2). The key component is HT in the top product. Similar to the Column (1), the concentration of HT is extremely low, and the concentrations of DT and T₂ are negligible in comparison with that of HT.

Hence, the ion chamber is appropriate to measure the concentration of HT in the top product of the Column (2).

To control the flow rate of the feed back stream, it is required to know the concentrations of HT in the external feed and side streams. From the simulation results, the concentrations of HT in these streams are expected to be in the range of 1~20%. The concentrations of DT and T₂ are larger than that of HT in these streams. The raman method is the most appropriate as the analysis instrument for measurement of HT concentrations.

3.2.3 Column (3)

This column has a function of production of high purity D₂. The top product is prior, and the Mode 1 is appropriate for the column (3). The HD, HT, and D₂ are recovered at the top while DT and T₂ are recovered at the bottom. The key component is DT in the top product. The concentration of HT is comparative to that for DT as seen in Table 1. The concentration of DT should be measured without influence of existence of HT. The combination of the ion chamber and the gas chromatograph is the only analysis instrument that satisfies this condition.

The design concepts for the control system of the columns are summarized in Table 5.

3.3 Proportional-integral control mode for the cryogenic distillation column

Kinoshita et al. [15] reported that the feedback control is appropriate for a cryogenic distillation column. They applied the proportional integral action as the control mode and proposed a parameter setting method of the control mode. In this

section, the parameters of the proportional-integral control mode were chosen for a representative case of column (3) in accordance with the method proposed by them. Stability of the control mode is also discussed for the representative case.

3.3.1 Parameter setting method [15]

The assumed upset is a step increase of 10% in the molar flow rate of D-T fed to the column. To keep the total feed flow rate, molar flow rate of D₂ is decreased for the corresponding value. Table 6 shows the assumed upset feed composition and the controlled and manipulated variables of the column. The top composition of column (3) in Table 6 differs from that in Table 2; this is because the computer code simulating dynamic behavior of the column does not consider the factors such as the nonideality of hydrogen isotope solutions and the heat balance between isotopic species. It is expected that the qualitative aspects of the conclusions described later are not vitiated at all even if the calculation is made by using a model neglecting these factors. The basic equations and calculation procedure of the code are also described in Appendix. The increase of molar flow rate of D-T in the feed stream by 10% is corresponding to that by ~6 mol/h. Hence, the value of the change in the manipulated variable needed for a successful control, ΔD , is expected to be about ~-6 mol/h. The response curves of the controlled variable were first calculated for the step changes in the manipulated variable ($\Delta D = -6$ and -10 mol/h). The response curves had no inflection points and hence the first-order lag system could be assured. Since the process transfer function is expected by the first-order lag system, the open loop transfer function is written by

$$G(s) = \frac{KM \left[1 + \frac{1}{T_i s} \right]}{1 + T_c s} \quad (2)$$

The characteristics equation is expressed by

$$T_i T_c s^2 + T_i(1 + KM)s + KM = 0 \quad (3)$$

In this case, the control never brings instability because the above equation has only real negative roots or imaginary roots whose real parts are negative. However, to obtain oscillatory behavior, it must have imaginary roots:

$$T_i \leq \frac{4KM T_c}{(1 + KM)^2} \quad (4)$$

The order of magnitude of K can be estimated from the following equation representing the proportional-integral action.

$$-\Delta D(t) = K \Delta y_{DT,1} + \frac{K}{T_i} \int_0^t \Delta y_{DT,1}(t) dt \quad (5)$$

The initial value of the controlled variable, $y_{DT,1}$, is $\sim 2.0 \times 10^{-4}$. It is desired that $\Delta y_{DT,1}$ be approximately one order of magnitude smaller than the initial value. The value of ΔD is in the range of ~ 5 mol/h. Hence, the approximate value of K is expected to be $\sim 2 \times 10^5$ mol/h. Substituting this K value into Eq. (4), we can obtain the maximum value of T_i . If the measurement of the controlled variable is accompanied by a time lag, by τ (h), the open loop transfer function is written by

$$G(s) = KM \left[1 + \frac{1}{T_i s} \right] \frac{\exp(-\tau s)}{1 + T_c s} \quad (6)$$

The characteristics equation is then written by

$$T_i s(1 + T_c s) + KM(1 + T_i s) \exp(-\tau s) = 0 \quad (7)$$

Substituting $s=j\omega_c$ in the above equation yields the following nonlinear simultaneous equations:

$$T_{ic}\omega_c + KMT_{ic}\omega_c \cos(\tau\omega_c) - KM \sin(\tau\omega_c) = 0 \quad , \quad (8)$$

$$-T_{ic}T_c\omega_c^2 + KMT_{ic}\omega_c \sin(\tau\omega_c) + KM \cos(\tau\omega_c) = 0 \quad , \quad (9)$$

$$\tau\omega_c \leq \frac{\pi}{2} \quad (10)$$

If K , M , T_c , and τ are given; the lower limit of the integral time, T_{ic} , can be calculated by solving the above equations by use of the Newton-Raphson method where T_{ic} and ω_c are the independent variables.

3.3.2 Stability of control method

The parameters in Eq. (2) were determined from the response curves of the controlled variable. Since the value of K is in the range of 2×10^5 , the maximum T_i can be estimated from Eq. (4). Table 7 shows the values of parameters in Eq. (2) and the maximum T_i thus obtained. For the columns (1) and (2), the ion chamber is used as the analysis instrument to control the purity of top product. The ion chamber can give us an analysis result with a negligible short time lag. Hence, the control mode proposed in the present study never becomes unstable as revealed in Eq. (3). We can select small T_i .

For the Column (3), the gas chromatograph must be used as the analysis instrument (See Table 5). The gas chromatography needs appreciable time for the measurement of a sample. In the case where the measurement is accompanied by a time lag, there is the lower limit of T_{ic} as indicated in Eqs. (8)~(10). Figure 5 shows the curves of T_{ic} plotted against τ . A longer time lag requires a larger integral time to

avoid the instability. In addition, there is no solution of Eqs. (8)-(10) in the case where τ is greater than 0.8 h for $\Delta D = -6$ mol/h (0.85 h for $\Delta D = -10$ mol/h). The time lag in measurement, τ , must be smaller than 0.8h. The maximum value of T_1 is 0.46 h for $\Delta D = -6$ mol/h (See Table 6). The lower limit of T_1 reaches 0.4 h for $\Delta D = -6$ mol/h as shown in Fig. 5. For these limitations also, the value of τ must be smaller than 0.8 h. We must pay attention to further disadvantage of the gas chromatography that it can not continually give us the measurement results. For this case, Kinoshita has reported that the time lag for measurement must be considered to be 50% larger than actual time required for composition measurement[16]. For the above discussion, it can be concluded that the permissible time for the gas chromatography is about 0.5 h. It is possible to complete the measurement for a sample within 0.5 h by the gas chromatography [17]. Thus, our final conclusion is that the gas chromatography is the applicable analysis method for the control system of the columns. However, to increase the stability of the control system, we must improve the gas chromatography. The shorter measurement time brings more stable control. For the control of column (3), we need the concentration of DT alone (See Table 5). Although the concentration of HT is comparative to that of DT, T_2 is negligible. This means that complete separation is needed only for HT and DT peaks in the chromatograms. It would be possible to reduce the measurement time by increasing the flow rate of the carrier gas and by adjusting treatment method of packings of the gas chromatograph [17].

4. Conclusion

In the design of column cascades for the fuel cycle of fusion reactors, the following requirements should be considered: either top or bottom product is prior to the other; there is no recycle stream between the columns in the cascade; and it is avoided to withdraw side streams as products or feeds of down stream columns. The proposed cascade consists of three columns with a feedback stream and satisfies all these requirements.

The disturbances of the cascade are expected to be the changes of feed streams (flow rate and compositions) and HETP value for each theoretical stage. For the changes of NBI flow rate, concentration of H in the torus exhaust, and stage numbers of side and feed streams, the purity of product of the cascade can be maintained just by adjusting the top and bottom flow rates. It is thus proved that the cascade has great flexibility.

Two basic control modes have been designed for the columns with and without a feedback stream, where either top or bottom product was prior to the other. The key component in the product stream and analysis instruments of the key component, which were the ion chamber, raman, and gas chromatography, were selected for each column in the cascade.

The designed control system never brings instability as long as the concentration of the key component is measured with negligible time lag. The ion chamber and raman satisfy this condition. However, the gas chromatography needs appreciable time for the measurement of a sample. The time lag for the measurement considerably affects the stability of the control system. A significant conclusion is that permissible time for the measurement is about 0.5 hour. It is possible to complete the measurement within 0.5 hour by the gas chromatography. The analysis method using the gas chromatography is valid for the control of the columns.

NOMENCLATURE

- B flow rate of bottom stream (mol/h)
- D flow rate of top product (mol/h)
- ΔD magnitude of top product flow rate upset assumed (mol/h)
- d inner diameter of column (cm)
- F flow rate of feed stream (mol/h)
- H_L liquid holdup for a theoretical stage (mol)
- h height of column (m)
- K proportional sensitivity (mol/h)
- K_p liquid-vapor equilibrium constant (-)
- I_T inventory of tritium within column (g)
- L liquid flow rate within column (mol/h)
- M static gain (h/mol)
- N number of total theoretical stages (-)
- N_F feed stage number (-)
- N_S side stream stage number (-)
- P pressure (kPa)
- R reflux ratio (-)
- T temperature (K)
- T_c time constant for first-order lag system (h)
- T_i integral time (h)
- t time (h)
- V vapor flow rate within column (mol/h)
- W_S flow rate of side stream (mol/h)
- x mole fraction in liquid stream (-)
- y mole fraction in vapor stream (-)
- $y_{DT,1}$ mole fraction of DT in top product (-)

$\Delta y_{DT,1}$ magnitude of mole fraction of DT upset (-)

Z atom fraction in side stream (-)

z mole fraction in feed stream (-)

τ time lag in measurement of controlled variable (h)

ω angular frequency (1/h)

Subscript

c critical value

D deuterium

H hydrogen

i component

j stage

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Table 1 Design values of hydrogen isotope gases by ITER-EDA and CDA

System	EDA	CDA
Blanket	To be determined	105 mol/h 99%H, 1%H
NBI	0	378 mol/h 0.5%H, 98%D, 1.5%T
Pellet Injector	To be determined ^{*1}	397 mol/h 92%H, 4%D, 4%T
Plasma exhaust gas	160 mol/h ^{*2} 1%H, 49.5%D, 49.5%T	71.4 mol/h 1%H, 49.5%D, 49.5%T

*1: No gas is sent from PI to the isotope separation system, since the pellet injector has a self-cleaning system.

*2: Only 10 % of the plasma exhaust gas (16 mol/h) is sent to the isotope separation system to maintain the hydrogen concentration in the D-T fuel gas at a desired value.

Table 3 Effect of side stream location and feed flow rates on performance of column (2)

Disturbance	$N_F = 45$ and 65 $N_S = 54$	$N_F = 65$ and 85 $N_S = 74$	NBI flow rate is decreased from 100 to 50 mol/h.	Concentration of H in plasma exhaust is increased from 1 to 5%.
Change of operational conditions*	D=20.0 B=191.4	D=24.2 B=187.2	D=21.0 B = 140.4	D=24.4 B=187.0
Atom fraction of T in top stream	6.8×10^{-8}	7.9×10^{-8}	7.9×10^{-8}	9.2×10^{-8}
Mole fraction of HT in bottom stream	2.93×10^{-5}	9.32×10^{-5}	7.30×10^{-6}	6.16×10^{-5}

* Other operational conditions are identical to those in Table 1.

Table 2 Design and operation parameters of columns in the proposed cascade

Column	(1)	(2)	(3)
N	80	120	80
N _F	55	55 (F ₁) and 75 (F ₂)	55
N _S	--	64	--
F (mol/h)	405	140 (F ₁) and 71.4 (F ₂)	190.2
D (mol/h)	365	21.2	109
B (mol/h)	40	190.2	81.2
W _S (mol/h)	--	600	--
R	2.0	50	20
P (kPa)	101.3	101.3	101.3
d (cm)	6.5	6.6	9.9
h (m)	3.9	5.9	5.9
I _T (g)	15	35	162
Top composition	H ₂ : 0.9792 HD: 2.08x10 ⁻² HT: 1.12x10 ⁻⁷ D ₂ : 0.0 DT: 0.0 T ₂ : 0.0	H ₂ : 0.8558 HD: 0.1442 HT: 1.82x10 ⁻⁷ D ₂ : 0.0 DT: 0.0 T ₂ : 0.0	H ₂ : 0.0 HD: 2.65x10 ⁻⁵ HT: 9.98x10 ⁻⁵ D ₂ : 0.9992 DT: 6.93x10 ⁻⁴ T ₂ : 2.22x10 ⁻⁹
Bottom composition	H ₂ : 2.70x10 ⁻⁶ HD: 0.3513 HT: 0.5851 D ₂ : 1.42x10 ⁻² DT: 3.09x10 ⁻² T ₂ : 1.82x10 ⁻²	H ₂ : 0.0 HD: 1.52x10 ⁻⁵ HT: 5.72x10 ⁻⁵ D ₂ : 0.5779 DT: 0.3196 T ₂ : 0.1024	H ₂ : 0.0 HD: 6.7x10 ⁻¹⁰ HT: 2.45x10 ⁻⁹ D ₂ : 1.24x10 ⁻² DT: 0.7477 T ₂ : 0.2399

Table 4 Change of design specifications with product purity required

Case	1	2	3
Column	(3)	(3)	(4)
N	80	120	120
N _F	55	60	70
N _S	--	70	--
F (mol/h)	190.2	190.2	109
D (mol/h)	90.5	134.8	107
B (mol/h)	99.7	55.4	2
W _S (mol/h)	--	700	--
R	13	14	15
P (kPa)	101.3	101.3	101.3
d (cm)	7.4	9.3	8.5
h (m)	3.9	5.9	5.9
I _T (g)	54	329	1.5
Top composition	H ₂ : 0.0 HD : 3.19x10 ⁻⁵ HT : 1.20x10 ⁻⁴ D ₂ : 0.9992 DT : 6.23x10 ⁻⁴ T ₂ : 5.41x10 ⁻⁹	H ₂ : 0.0 HD : 3.45x10 ⁻⁵ HT : 6.77x10 ⁻⁵ D ₂ : 0.9992 DT : 6.86x10 ⁻⁴ T ₂ : 2.49x10 ⁻⁹	H ₂ : 5.03x10 ⁻⁹ HD : 1.29x10 ⁻⁴ HT : 4.62x10 ⁻⁸ D ₂ : 0.9999 DT : 3.81x10 ⁻⁸ T ₂ : 0.0
Bottom composition	H ₂ : 0.0 HD : 9.48x10 ⁻⁹ HT : 7.62x10 ⁻⁸ D ₂ : 0.1955 DT : 0.6092 T ₂ : 0.1954	H ₂ : 0.0 HD : 0.0 HT : 0.0 D ₂ : 1.48x10 ⁻⁵ DT : 0.2012 T ₂ : 0.7988	H ₂ : 0.0 HD : 0.0 HT : 0.0 D ₂ : 0.9568 DT : 4.32x10 ⁻² T ₂ : 9.05x10 ⁻⁶

The column (4) is added at the down stream of the column (3). The top product of column (3) is fed to the column (4) through an equilibrator.

Table 5 Design concept for control system of each column

Column	(1)	(2)	(3)
Important product	Top	Top	Top
Key component	HT	HT	DT
Control mode	Mode 1	Mode 2	Mode 1
Analysis method	Ion chamber	Ion chamber for top stream Raman for feed and side stream	Ion chamber + gas chromatography

Table 6 Controlled and manipulated valuables of column (3)

Disturbance	Increase of D-T flow rate in feed composition (10%)
Upset Feed composition	H ₂ : 0.0 HD : 1.52x10 ⁻⁵ HT : 5.72x10 ⁻⁵ D ₂ : 0.5459 DT : 0.3516 T ₂ : 0.1024
Top composition	H ₂ : 0.0 HD : 2.65x10 ⁻⁵ HT : 9.98x10 ⁻⁵ D ₂ : 0.9997 DT : 1.31x10 ⁻⁴ T ₂ : 1.1x10 ⁻¹⁰
Controlled	Mole fraction of D-T in top product (y _{DT,1} ~2.0x10 ⁻⁴)
Manipulated	Flow rate of top product

Table 7 Optimum values of parameters in transfer function and proportional-integral controller

ΔD (mol/h)	T _c (h)	M (h/mol)	K (mol/h)	Maximum Ti (h)
- 6	0.47	4.0x10 ⁻⁶	2x10 ⁵	0.46
- 10	0.70	5.0x10 ⁻⁶	2x10 ⁵	0.70

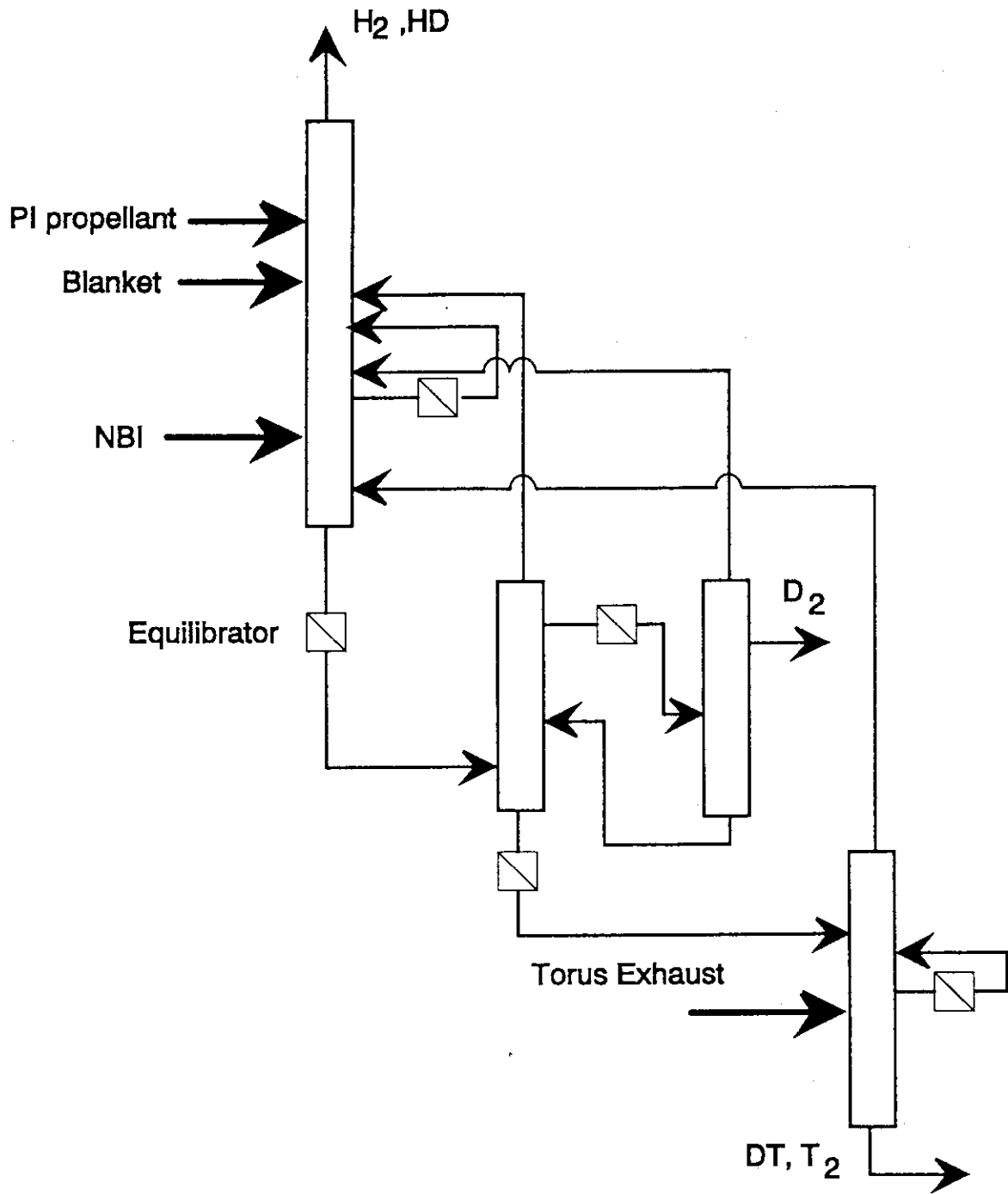


Figure 1 Column cascade proposed by Sze et al [5].

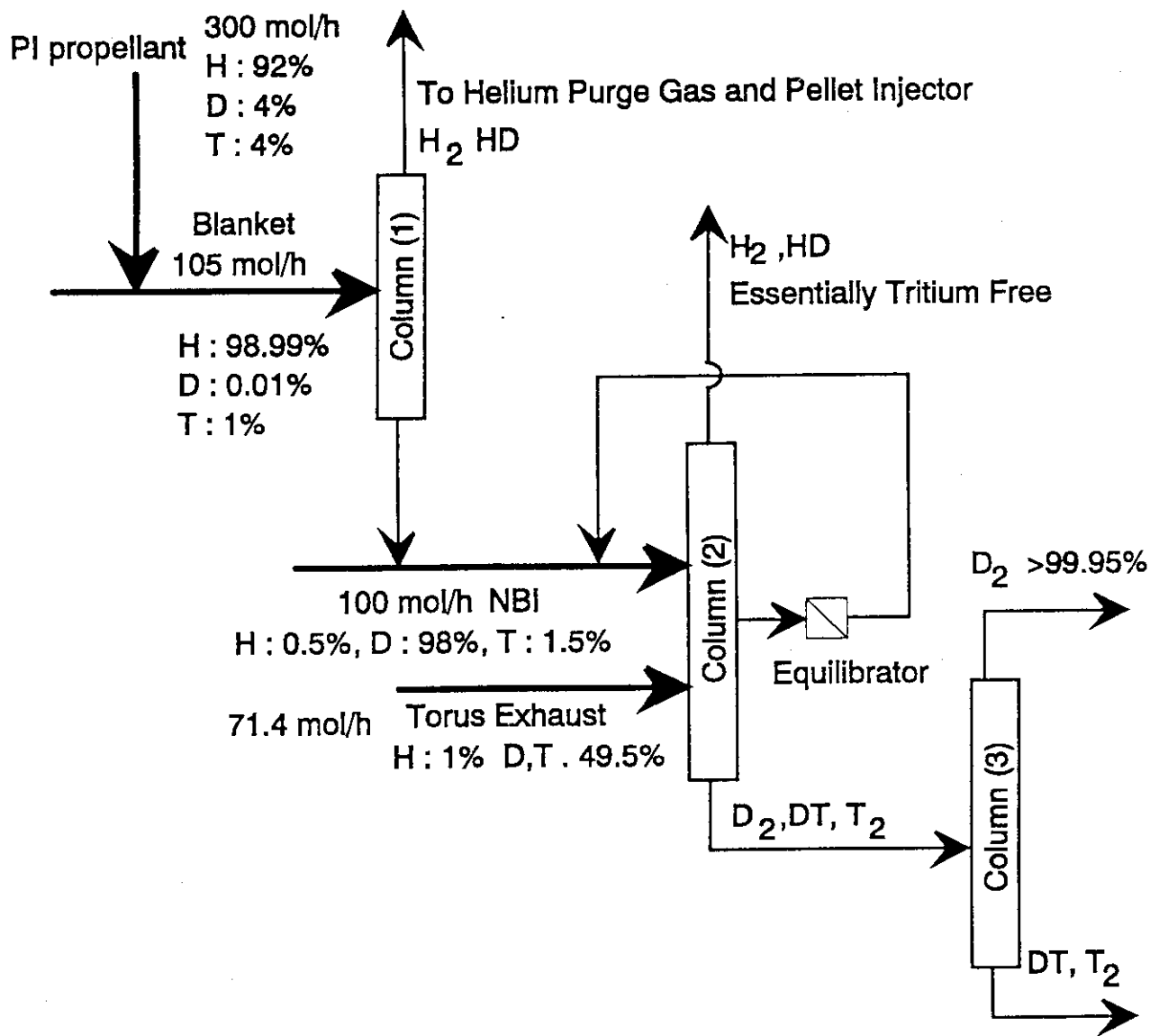


Figure 2 Column cascade proposed in the present study.

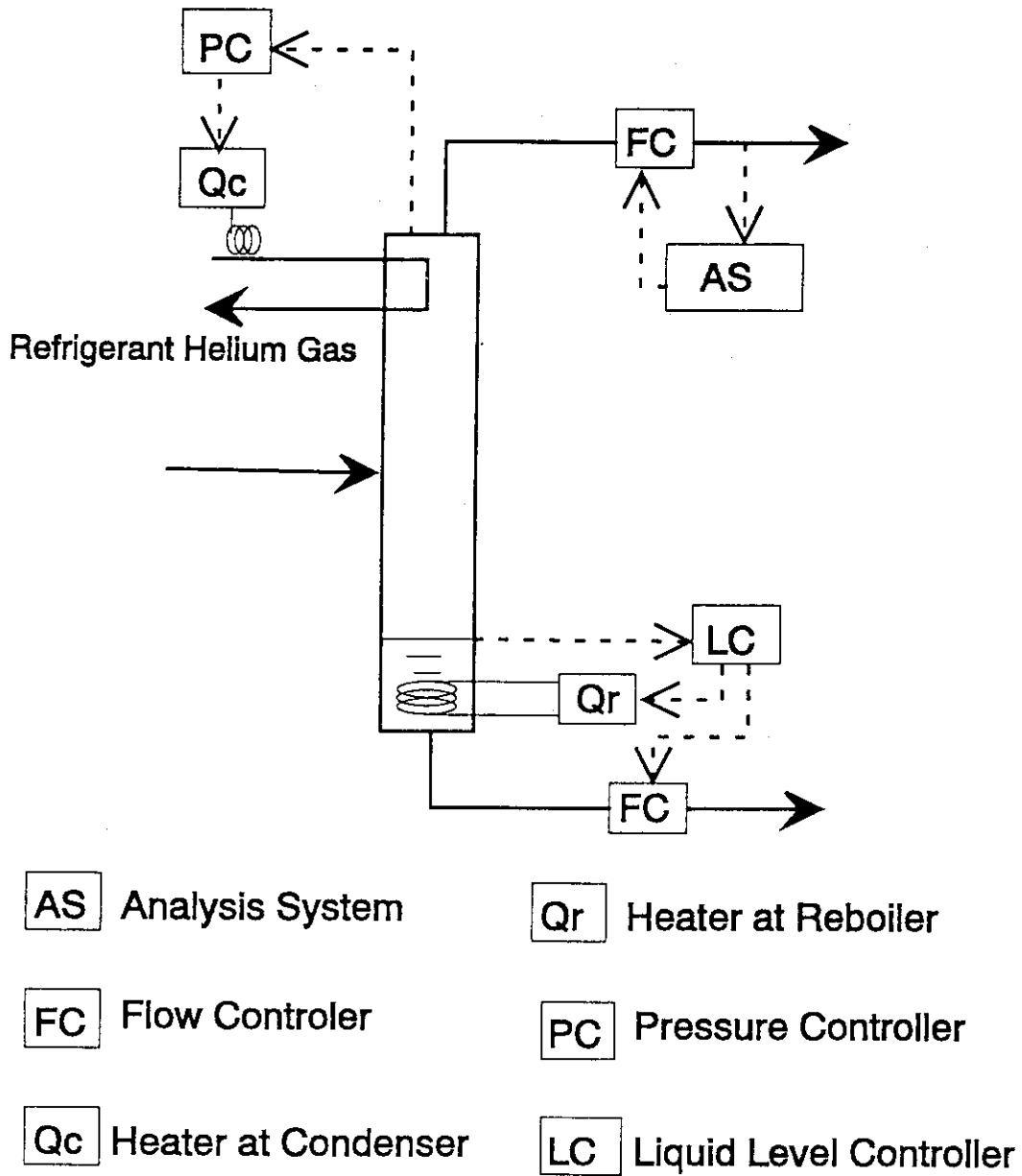


Figure 3 Typical control system for the column whose top product is more important than bottom product.

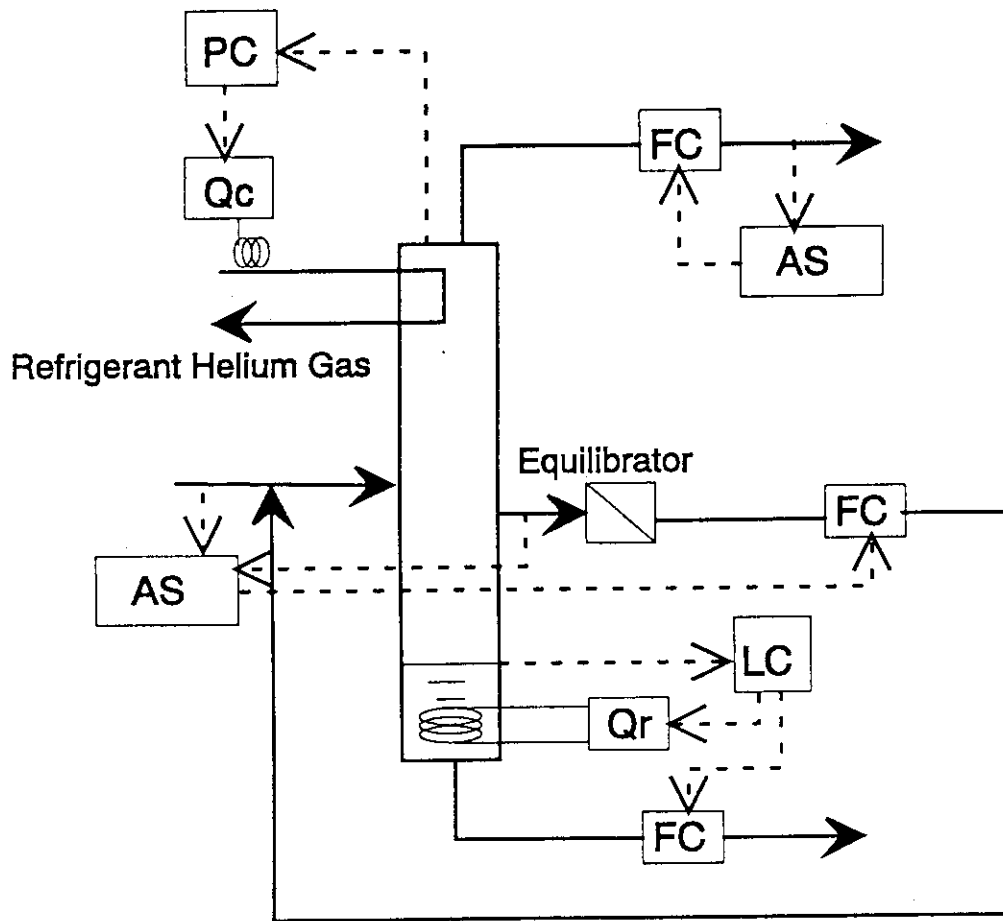


Figure 4 Control system for the column having a feed back stream.

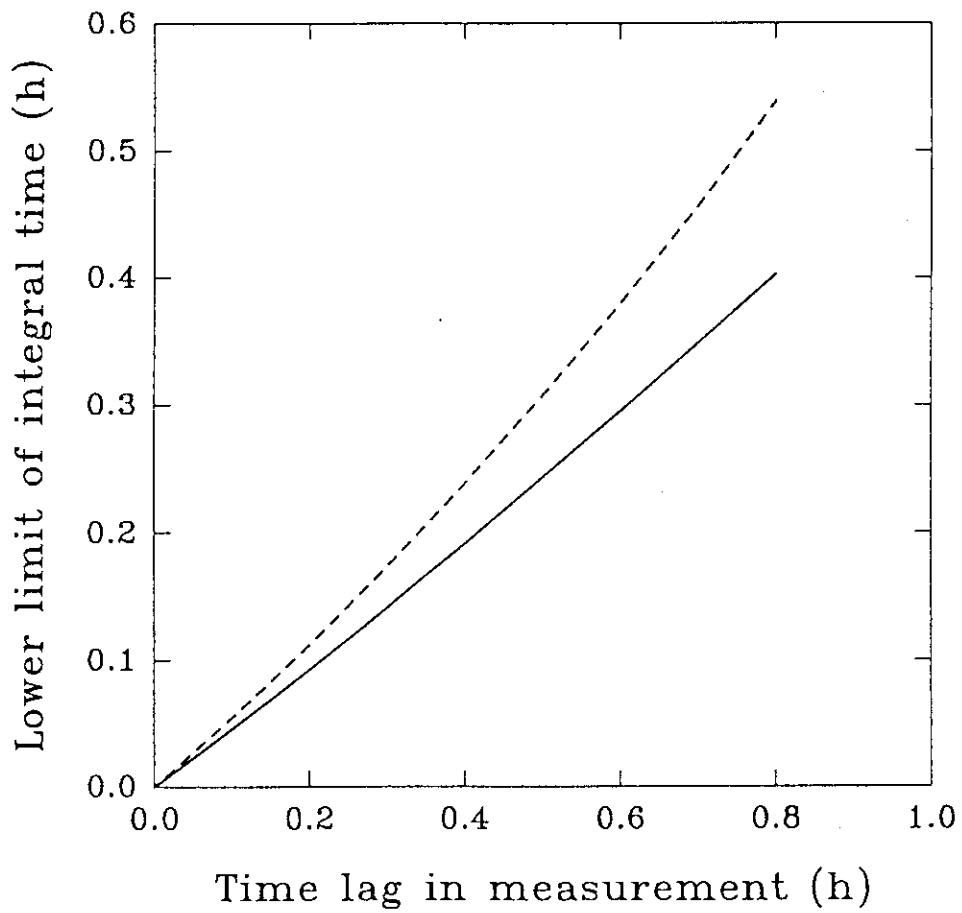


Figure 5 Curve of lower limit of integral time plotted against time lag in measurement of controlled variable; —, $\Delta D = -6$ mol/h; ---, $\Delta D = -10$ mol/h.

Appendix Mathematical simulation procedure for columns with and without a feedback stream

If the atom fraction of hydrogen in the side stream, Z_H , and that of deuterium, Z_D , are given, the composition of the stream fed to the column can be determined. Then, the composition distribution within the column is computed by main distillation calculation. The calculated values of the atom fraction of hydrogen 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 [18]:

$$f_H(Z_H, Z_D) = Z'_H - Z_H = 0, \quad f_D(Z_H, Z_D) = Z'_D - Z_D = 0 \quad (11)$$

Equation (11) can be solved by the two-dimensional Newton-Raphson method:

$$Z_k^{i+1} = Z_k^i + \Delta Z_k^i, \quad (12)$$

where

$$\begin{bmatrix} \partial f_H / \partial Z_H & \partial f_H / \partial Z_D \\ \partial f_D / \partial Z_H & \partial f_D / \partial Z_D \end{bmatrix} \begin{bmatrix} \Delta Z_H \\ \Delta Z_D \end{bmatrix} = - \begin{bmatrix} f_H \\ f_D \end{bmatrix} \quad (13)$$

The superscript, i , denotes the iteration number. The partial derivative is numerically calculated by

$$\left(\partial f_j / \partial Z_k \right) \cong \frac{f_j(Z_k + \Delta Z_k) - f_j(Z_k - \Delta Z_k)}{2\Delta Z_k} \quad (14)$$

The main distillation calculation is performed by finding out the solutions of the following N -dimensional nonlinear simultaneous equations:

$$\left. \begin{array}{l} S_1(T_1, \dots, T_N) = 0 \\ \dots \\ S_N(T_1, \dots, T_N) = 0 \end{array} \right\} , \quad (15)$$

where S_j is defined by

$$S_j = \sum_{i=1}^m K_{p_{i,j}} x_{i,j} - 1 \quad (16)$$

The liquid compositions, $x_{i,j}$, are obtained by solving the following equation.

$$\begin{array}{cccccc} \left\| \begin{array}{ccccc} B_{i,1} & C_{i,1} & 0 & \dots & \dots \\ A_{i,2} & B_{i,2} & C_{i,2} & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots \\ & A_{i,j} & B_{i,j} & C_{i,j} & \\ 0 & \dots & \dots & A_{i,N} & B_{i,N} \end{array} \right\| \cdot \begin{array}{c} \left\| \begin{array}{c} x_{i,1} \\ x_{i,2} \\ \cdot \\ x_{i,j} \\ x_{i,N} \end{array} \right\| = \begin{array}{c} \left\| \begin{array}{c} D_{i,1} \\ D_{i,2} \\ \cdot \\ D_{i,j} \\ D_{i,N} \end{array} \right\| \end{array} \quad (17)$$

where

$$A_{i,j} = L_{j-1}, \quad B_{i,j} = -(V_j + W_{S,j})K_{p_{i,j}} - L_j, \quad C_{i,j} = V_{j+1}K_{p_{i,j+1}}, \quad D_{i,j} = -F_j z_{i,j}$$

Equation (15) is solved by application of the modified Newton-Raphson method [16].

The new values of the independent variables for the $(\beta+1)$ 'th iteration are calculated from

$$T_j^{\beta+1} = T_j^{\beta} + \Delta T_j^{\beta} \quad (18)$$

where

$$\begin{bmatrix} \partial S_1 / \partial T_1 & \dots & \partial S_1 / \partial T_N \\ \dots & \dots & \dots \\ \partial S_N / \partial T_1 & \dots & \partial S_N / \partial T_N \end{bmatrix} \begin{bmatrix} \Delta T_1^\beta \\ \dots \\ \Delta T_N^\beta \end{bmatrix} = - \begin{bmatrix} S_1 \\ \dots \\ S_N \end{bmatrix} \quad (19)$$

To consider all the factors: (i) nonideality of hydrogen isotope solution; (ii) differences in the latent heat of vaporization among the hydrogen isotopes; and (iii) decay heat of tritium, liquid flow rate within the column and activity coefficients of the solutions are recalculated in an outer loop of the main distillation calculation.

For the dynamic simulation, basic equations are expressed by

$$H_{Lj} \frac{dx_{i,j}}{dt} = V_{j+1}y_{i,j+1} + L_{j-1}x_{i,j-1} - L_jx_{i,j} - (V_j + W_{S,j})y_{i,j} + F_jz_{i,j} \quad (20)$$

The Improved Euler method was used to solve this equation because of its simple algorithm.