

Computer Program Development for D₂O Upgrader Performance Management

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중수승급기 성능관리 프로그램 개발

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Abstract

Heavy water is used as a moderator and a coolant in the pressurized heavy water reactor. Because of the high cost of heavy water, downgraded heavy water generated in the reactor system is recycled to the reactor after being concentrated up to 99.8% or more in heavy water upgraders. This study investigates the process of upgraders and then suggests a theoretical model. The relations between process variables are derived from tower packing characteristics, vapour-liquid equilibria, and mass-heat balance equations at a steady state operation of the upgrader. A computer program UPGR is developed, using the algorithm that solves the nonlinear equations step by step. It shows that the results of computer simulation are in good agreement with the operating data of the Wolsung upgrader. Thus, this computer code offers the optimum operating guide and is now applied to manage the performance of upgraders for the effective operation of the heavy water upgraders.

요 약

중수는 중수형 원자로의 감속재 및 냉각재로 사용되고 있으며 그 가격이 고가이기 때문에

일단 계통내에서 사용된 후 농도가 낮아진 저등급 중수는 중수승급기를 통해 99.8% 이상으로 농축 재생되어 중수로로 재주입되고 있다.

본 연구에서는 중수승급기의 공정을 면밀히 검토하였고 정상상태의 중수증류공정의 해석을 위하여 이론적인 모델을 제시하였으며 변수들간의 관계식을 설정하였다. 그리고 이 비선형 관계식을 단계적으로 처리하는 알고리즘의 전산 프로그램 UPGR을 개발하였다. 전산코드의 결과는 실제 운전 데이터와 잘 일치하였다. 월성 1호기에서 이를 이용한 운전지침의 제시, 운전 효율의 평가, 성능평가 및 성능관리를 수행함으로써 중수승급기의 효율적인 운전에 기여하고 있다.

Nomenclature

c	total number of components	
F	flow rate of feed stream	(kmol/hr)
G	gas load	(kg/m ² /hr)
H	enthalpy of liquid stream	(kcal/kmol)
h	enthalpy of vapour stream	(kcal/kmol)
hf	enthalpy of feed stream	(kcal/kmol)
i	component i	
j	stage j	
K _p	equilibrium constant for isotopic exchange reaction	
L	flow rate of liquid stream	(kmol/hr)
M	molar mass	(kg/kmol)
n	total number of theoretical stages	
P	pressure	(mbar)
Q	rate of heat transfer	(kcal/hr)
T	temperature	(°C)
L	flow rate of liquid stream	(kmol/hr)
V	flow rate of vapour stream	(kmol/hr)
vg	gas velocity	(m/sec)
x	mole fraction in liquid stream	
y	mole fraction in vapour stream	
z	mole fraction in feed stream	
π	vapor pressure	(mbar)
ρ _g	density of gas	(kg/m ³)

I. Introduction

The pressurized heavy water reactor, which is called CANDU, is being operated at Wolsung Nuclear Power Plant, Unit 1 in Korea. Heavy water is used as

a moderator and a coolant in the reactor. Because of the high cost of heavy water, downgraded heavy water generated in the reactor system is recycled to the reactor after being concentrated up to 99.8% or more in heavy water upgraders.[1] Figure 1 shows the heavy water cycle in the CANDU system.[2]

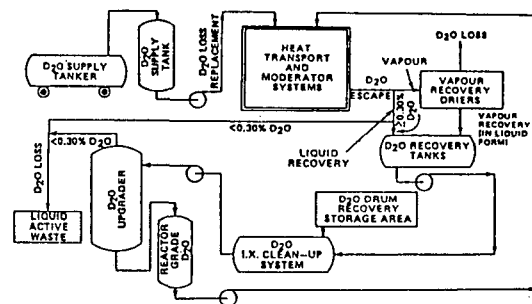


Fig. 1. Heavy Water Cycle in CANDU Systems

Water distillation is employed in the heavy water upgraders. Since the separation factor ($\alpha = 1.04$) is extremely small, a large number of theoretical stages are required.[3,4,5] Thus, high performance packing and vacuum distillation process, which are different from the general distillation process, are adopted in order to reduce the height of a tower.

Effective operation of the upgraders is indispensable to the nuclear power plant of the pressurized heavy water reactor. Thus, the appropriate description of physical phenomena in the reactor is an essential task for the its optimum design and operation.

In this work, the process of upgraders is investigated and a theoretical model is suggested. The relations between process variables are derived from tower packing characteristics, vapour-liquid equilibria, and

mass-heat balance equations for the analysis of the heavy-light water distillation process at a steady state. A computer code using the algorithm that solves the nonlinear equations step by step is also developed. The results obtained by computer simulation are in good agreement with the operating data of the upgrader of the Wolsung nuclear power plant. Thus, this computer code offers a good guide for the operation of upgrader and provides for the optimum performance of the heavy water upgrader through its effective operation.

II. Process Description and Packing Characteristics

The Wolsung station employs two heavy water upgraders, one for the moderator(Mod) system and the other for the primary heat transport(PHT) system. From the operational points of view, there is a significant advantage in having two upgraders since the Mod system has different water chemistry and tritium levels from the PHT system and the two upgraders permit independent operation of the two systems.[1]

Figure 2 shows a schematic representation of a Wolsung upgrader.[6] The upgrader would be approx-

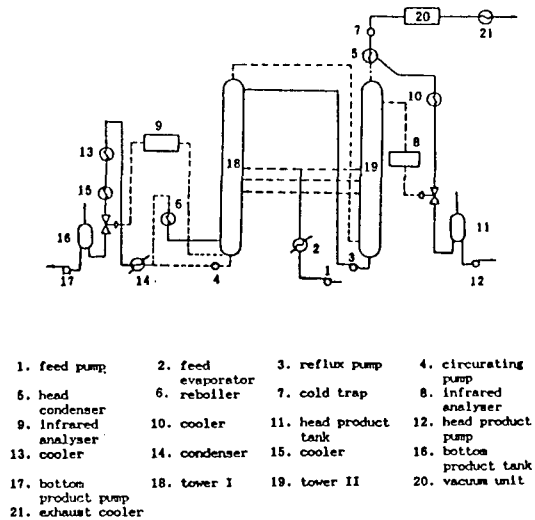


Fig. 2. Schematic Diagram of a Wolsung Upgrader

imately 70 meters high if it is a single column. To overcome this difficulty the large upgrader is divided into

two towers which are connected by two flow streams, one of a vapour and the other of a liquid reflux line. Heat is applied to the bottom of the first column to boil the liquid by a partial reboiler and is taken out at the top of the second column to condense the vapour by a total condenser. As the vapour stream rises in the column, it constantly transfers D₂O to the liquid stream and the liquid constantly transfers H₂O to the vapour stream. Thus, the D₂O content of the liquid stream increases as it falls through the tower. Feed in the form of saturated vapour from the feed evaporator enters into the tower through one or two of many feed points. The feed point is selected to match the concentration of the feed with that of the vapour in the tower. In order to obtain optimum performance of the upgrader, it is usually operated at sub-atmospheric pressure (approximately 13.2 kPa at the top). Thus, a vacuum system is required to remove both air from the towers before start-up and non-condensibles during operation. To prevent escape of heavy water vapour cold traps are provided in the vacuum system.

A tower section contains several layers of Sulzer CY packing, a collector, and a distributor. The PHT and Mod upgraders has 20 and 17 tower sections, respectively. This packing has been developed for heavy water rectification by Sulzer Co.[7] It consists of parallel, corrugated strips of Cu-Sn wire mesh (Fig. 3), and has a hydraulic diameter of 6mm and 700 m²/m³ exchange surface.[7] The main advantage of this packing is lowering the height of the column requiring a large number of theoretical plates.[8] In general, the number

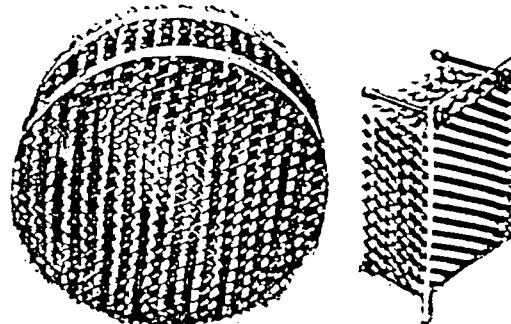


Fig. 3. The Structure of Sulzer CY Packing

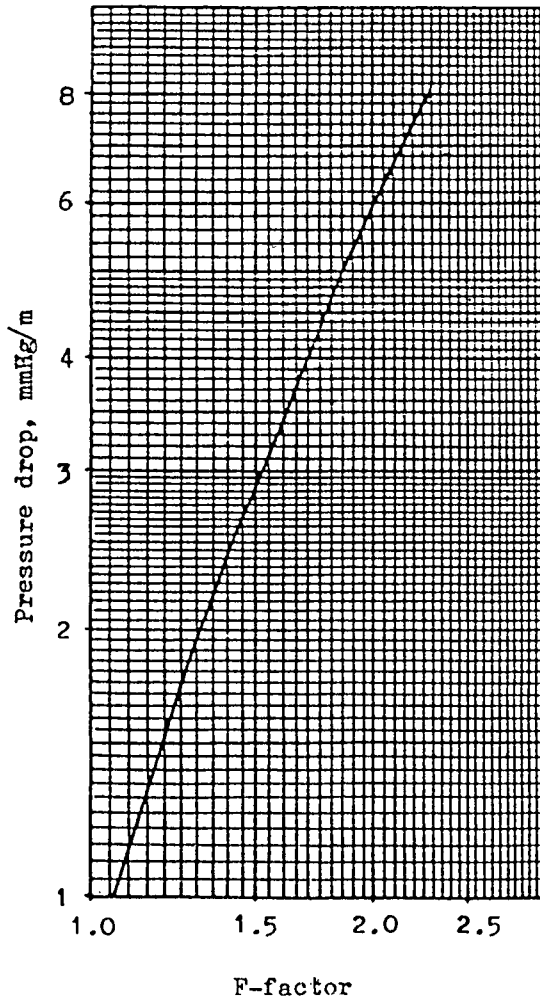


Fig. 4. Pressure Drop of Sulzer CY Packing as a Function of F-factor

of theoretical stages per meter (NTSM) of a packing depends on velocity, density, and pressure of vapour stream. NTSM of this CY packing is 8-12 at the operating conditions of the upgraders.

Fig. 4 shows the pressure drop per unit height of the CY packing for different gas loads. This is an important parameter for the analysis of distillation column. The gas load is often expressed as the F-factor, which is defined as follows.[8]

$$F\text{-factor} = v_g \sqrt{\rho_g} = 0.002533G \sqrt{\frac{T}{M \cdot P}}$$

where v_g : gas velocity, m/sec

ρ_g : gas density, kg/m³

T : temperature, °C

M : molar mass, kg/kmol

P : pressure, mbar

Equipment dimensions and typical operating conditions of the two upgraders are summarized in Table 1. The process of Mod upgrader is the same as that of PHT upgrader except for the number of feeds. The PHT upgrader requires a single feed, while the Mod upgrader uses a single or dual feed.

III. Theoretical Analysis

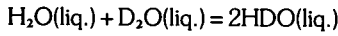
III-1. Vapor-liquid equilibria and atomic fraction

The mixture of heavy and light water is composed of D₂O, HDO, and H₂O inevitably and the isotopic ex-

Table 1. Equipments dimensions & operating conditions of upgraders

	PHT	Mod
Tower I	0.8m ϕ \times 36.4mH	0.8m ϕ \times 33.5mH
Tower II	0.8m ϕ \times 38.2mH	0.8m ϕ \times 33.2mH
Tower I + II	0.8m ϕ \times 74.6mH	0.8m ϕ \times 66.7mH
Total packed height	43.36m	38.56m
No. of sections	20	17
No. of feed points	16	13
Bottom concentration	>99.8 wt % D ₂ O	>99.9 wt % D ₂ O
Head concentration	< 0.1 wt % D ₂ O	< 0.1 wt % D ₂ O

change reaction is as follows.[9]



The reaction rate is known to be very rapid[10] and the equilibrium constant K_p can be expressed by

$$K_p = x(HDO)^2 / \{x(H_2O) x(D_2O)\} \quad (1)$$

where x denotes liquid mole fraction. The constant K_p , which is a function of the temperature in degrees centimeter only, was given empirically by I. Kirshenbaum as follows.[10]

$$K_p = 3.7621 + 1.5057E-03T - 4.0E-06 T^2$$

If we assume that Raoult's law is applicable to the mixture and the vapour pressure of HDO is the geometric mean of those of H_2O and D_2O , that is, $\pi(HDO) = \sqrt{\pi(H_2O)\pi(D_2O)}$, then the equilibrium constant can be rewritten as[11, 12, 13]

$$K_p = \{y(HDO) P/\pi(HDO)\}^2 / \{ \{y(D_2O) P/\pi(D_2O)\} \{y(H_2O) P/\pi(H_2O)\} \} \\ = y(HDO)^2 / \{y(H_2O)y(D_2O)\} \quad (2)$$

Here, P is the pressure of the system and y is vapour mole fraction.

If the temperature and the pressure of the system are known, mole fractions of liquid and vapour phase can be calculated from the following Eqs. (3)-(6).

$$x(H_2O) + \sqrt{K_p} x(H_2O) x(D_2O) + x(D_2O) = 1 \quad (3)$$

$$x(H_2O)\pi(H_2O) + \sqrt{K_p} x(H_2O)\pi(H_2O) x(D_2O)\pi(D_2O) + x(D_2O)\pi(D_2O) = P \quad (4)$$

$$y(H_2O) = \pi(H_2O) x(H_2O)/P \quad (5)$$

$$y(D_2O) = \pi(D_2O) x(D_2O)/P \quad (6)$$

In Eqs. (3)-(6), independent variables are temperature, pressure, $x(H_2O)$, $x(D_2O)$, $y(H_2O)$, and $y(D_2O)$. In other word, if two of six variables are given the others can be calculated.

Mass and physical properties (e.g. enthalpy) of HDO cannot be measured, because HDO always exists in the H_2O and D_2O mixture. Atomic fractions x, y of hydrogen isotopes are defined as follows for simplicity of separation performance equations.

$$x(H) = x(H_2O) + x(HDO)/2, \\ y(H) = y(H_2O) + y(HDO)/2 \\ x(D) = x(D_2O) + x(HDO)/2, \\ y(D) = y(D_2O) + y(HDO)/2 \quad (7)$$

Here, HDO content is assumed to be equally present

in H_2O and D_2O content. Thus, energy and material balances are written successfully by the atomic fractions.[11, 14]

III-2. Theoretical model

A theoretical model of heavy water upgraders is illustrated in Fig. 5. The figure shows n theoretical stages arranged in a countercurrent cascade, where the stages are numbered down from the top, and a condenser and a reboiler are located at stage 1 and stage n , respectively. Heat is removed at a rate of $Q(1)$ from stage 1. On the other hand, heat is added at a rate of $Q(n)$ from the reboiler. It is assumed that phase equilibrium and isotopic exchange reaction equilibrium are achieved instantly in the column and no heat transfer through the column occurs.

A schematic representation of an equilibrium stage j is shown in Fig. 6. Saturated vapour-phase of molal flow rate $F(j)$ is fed into the stage j with properties of mole fractions $z(i, j)$ of component i , temperature $T(j)$, pressure $P(j)$, and overall molal enthalpy $hf(j)$, which

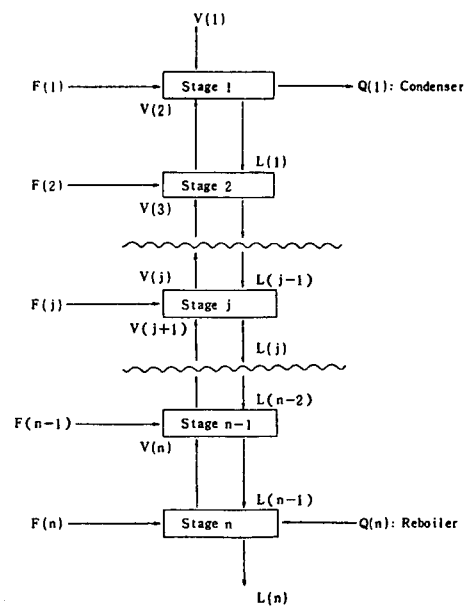


Fig. 5. Countercurrent Cascade of n Stages

are equal to those of vapour stream $V(j)$. The interstage liquid of molal flow rate $L(j-1)$ also enters the stage j from the stage $j-1$ above, with properties of mole fractions $x(i, j-1)$, enthalpy $H(j-1)$, temperature $T(j-1)$, and

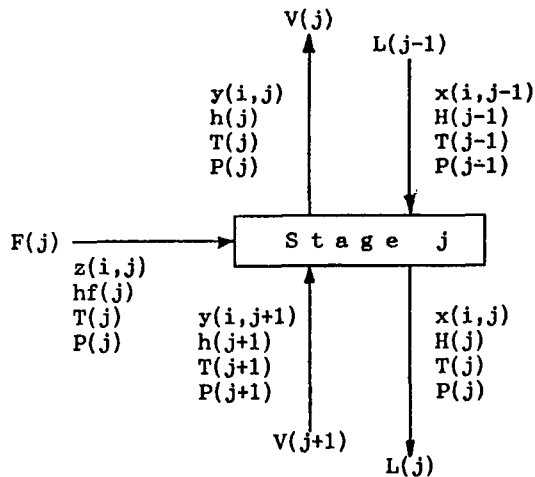


Fig. 6. A Schematic Representation of an Equilibrium Stage j

pressure $P(j-1)$, which is less than the pressure of stage j . Similarly, from the stage $j+1$ below, the interstage vapour of molal flow rate $V(j+1)$ enters the stage j , with properties of mole fractions $y(i, j+1)$, enthalpy $h(j+1)$, temperature $T(j+1)$, and pressure $P(j+1)$.

The interstage vapour of molal flow rate $V(j)$ leaves the stage j with intensive properties $y(i, j)$, $h(j)$, $T(j)$, and $P(j)$. The interstage liquid of molal flow rate $L(j)$ also leaves the stage j with intensive properties $x(i, j)$, $H(j)$, $T(j)$, and $P(j)$. These two exit streams are assumed to be in phase equilibrium.

III-3. Simulation procedure

Associated with each theoretical stage, the following equations can be written in terms of the variables set in Figs. 5 and 6 [15, 16].

- o Material balance for each component

$$L(j-1) x(i, j-1) + V(j+1) y(i, j+1) + F(j) z(i, j) - L(j) x(i, j) - V(j) y(i, j) = 0 \quad (8)$$

- o Phase equilibrium relation for each component

$$x(i, j) = \text{function of } \{P(j), T(j), y(1, j), \dots, y(c, j)\} \quad (9)$$

- o Mole fraction summations

$$\sum_{i=1}^c x(i, j) = 1, \quad \sum_{i=1}^c y(i, j) = 1 \quad (10)$$

- o Energy balance

$$L(j-1) H(j-1) + V(j+1) h(j+1) + F(j) hf(j) - L(j) H(j) - V(j) h(j) = 0 \quad (11)$$

- o Total material balance

$$L(j) = V(j+1) + \sum_{m=1}^j F(m) - V(1) \quad (12)$$

- o Pressure drop

$$P(j) = P(j-1) + \Delta P(j-1) \quad (13)$$

The equilibrium relation Eq. (9) is a condensed form of Eqs. (3)-(6) previously described, and the pressure drop for Eq. (13) which depends on the conditions of vapour stream can be obtained from Fig. 4. The subscript of component i can be omitted by denoting deuterium atomic fraction by x , since the three-component system can be reasonably handled as a binary system.

When molal flow rate F , compositions of feed z , top product $y(1)$ and bottom product $x(n)$, boil up rate V_{spec} , and head pressure $P(1)$ are specified, the number of theoretical stages n and all $x(j)$, $y(j)$, $L(j)$, $V(j)$, $T(j)$, and $P(j)$ can be calculated by solving the set of nonlinear equations above. The simulation starts from reading data and assuming the vapour flow rate $V(2)$ of stage 2. The procedure can be summarized as follows.

- (a) $x(1)$, $V(1)$, $T(1)$ of stage 1 (condenser) are calculated from the following equations.

$$x(1) = y(1)$$

$$V(1) = \sum_{j=1}^n F(j) \{z(j) - x(n)\} / \{y(1) - x(n)\} : \text{total material balance}$$

material balance

$$T(1) = \text{function of } \{P(1), y(1)\} : \text{equilibrium equation}$$

- (b) $y(2)$, $P(2)$, $T(2)$, $x(2)$ of stage 2 are obtained from the equations below.

$$y(2) = x(1), \quad P(2) = P(1) + \Delta P(1)$$

$$\{T(2), x(2)\} = \text{function of } \{P(2), y(2)\} : \text{equilibrium}$$

equation

- (c) The following equations with unknown variables of molal flow rates, enthalpies and pressure at the top stage are set.

$$L(1) + V(3) + F(2) - L(2) - V(2) = 0$$

$$L(1) x(1) + V(3) y(3) + F(2) z(2) - L(2) x(2) - V(2) y(2) = 0$$

$$L(1) H(1) + V(3) h(3) + F(2) hf(2) - L(2) H(2) - V(2) h(2) = 0$$

$$L(2) = V(3) + \sum_{m=1}^2 F(m) - V(1)$$

$$P(3) = P(2) + \Delta P(2) \quad (14)$$

Here, enthalpy H and h are functions of temperature T and compositions x any y . Also pressure drop ΔP is a function of vapour stream conditions. Thus, the unknown variables $L(1)$, $T(3)$, $V(3)$, and $L(2)$ in Eq. (14) are determined from the assumed variable $V(2)$. At arbitrary stage j , these equations can be written in the same manner.

$$L(j-1) + V(j+1) + F(j) - L(j) - V(j) = 0$$

$$L(j-1)x(j-1) + V(j+1)y(j+1) + F(j)z(j) - L(j)x(j) - V(j)y(j) = 0$$

$$L(j-1)H(j-1) + V(j+1)h(j+1) + F(j)hf(j) - L(j)H(j) - V(j)h(j) = 0$$

$$L(j) = V(j+1) + \sum_{m=1}^j F(m) - V(1)$$

$$P(j+1) = P(j) + \Delta P(j) \quad (15)$$

- (d) Assume $V(2)$

- (e) Assume $T(3)$

- (f) $L(1)$, $y(3)$, $V(3)$, and $L(2)$ are calculated from Eq. (14)

- (g) $P(3)$ is obtained from Fig. 4 by using F-factor calculated from the conditions of $V(2)$.

- (h) $T(3)$, $x(3)$ are calculated from

$$\{T(3), x(3)\} = \text{function of } \{P(3), y(3)\}: \text{equilibrium equation}$$

The calculated value $T(3)$ may not equal that of step (e), because the latter is an assumed value.

The iteration method is employed to match these two values within a certain error range and to find the solution of Eq. (14).

- (i) $L(1)$, $T(3)$, $y(3)$, $V(3)$, and $L(2)$ are calculated in step (e) through (h) by changing $T(3)$ value repeatedly until the variables satisfy Eq. (14) simultaneously.

- (j) $L(j-1)$, $T(j+1)$, $y(j+1)$, $V(j+1)$, and $L(j)$ are calculated sequentially from $j=3$ to $j=n$ by using Eq. (15). Step (e) through (i) are repeated sequentially by assuming $T(j+1)$ at the stage j , until the calculated value of $x(n)$ is equal to the specified value. During this step molal flow rates of feed $F(j)$ are determined according to composition of vapour stream $y(j)$. It is found that the calculated value of boil up rate $V(n)$ at the stage $n-1$ is different from the specified value in most cases. We denote the former by V_{cal} and the latter by V_{spec} .

- (k) The simulation program is considered to be solved, if the following convergence criterion is satisfied.

$$|V_{spec} - V_{cal}| < \text{err}, \quad 0 < \text{err} < 1$$

Step (d) through (j) are carried out repeatedly by changing $V(2)$ value. This iteration is performed by using the half-interval method, where $V(2)$ is the iterative variable.

The algorithm for the simulation is shown in Fig. 7.

Output variables are the number of theoretical stages and $x(j)$, $y(j)$, $L(j)$, $V(j)$, $T(j)$, and $P(j)$ for all j stage.

If other variables are specified, corresponding substitutions are made for output variables. For instance, if the number of theoretical stages is specified, molal flow rate of feed is substituted for the output variable. Thus, regardless of specifications, the simulation program can be solved by the iterative technique.

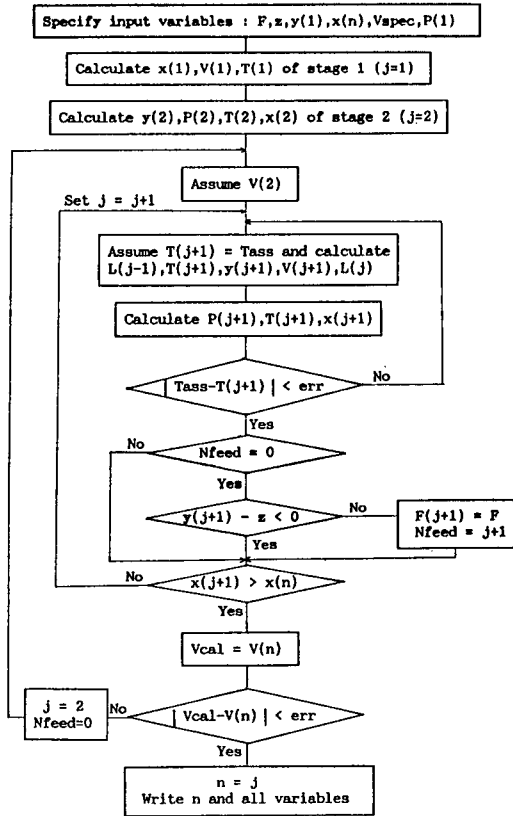


Fig. 7. Flow Chart of Simulation Procedure

IV. Result and Discussion

IV-1. The validity of the computer program

The computer program UPGR is written in FORTRAN language and executed in double precision.

At the operation of the PHT upgrader, the process variables including head concentration and pressure, bottom concentration, and boil up rate are maintained almost invariably as Table 2. Feed concentration changes very broadly, 0.5-99.5 wt % of D₂O. The flow rate of feed is closely related to its concentration for the upgrader whose number of theoretical stages is 505. It is assumed that feed enters stage j where the concentration of vapour stream is about the same as that of feed since feed enters into the tower through

Table 2. Process Variables of Wolsung D₂O Upgraders

	PHT	Mod	Unit
No. of theoretical stages	505	440	
Head concentration	0.017	0.017	wt % D ₂ O
Head pressure	13.2	13.2	kPa
Bottom concentration	99.83	99.91	wt % D ₂ O
Boil up rate	46.0	47.0	kmol/hr

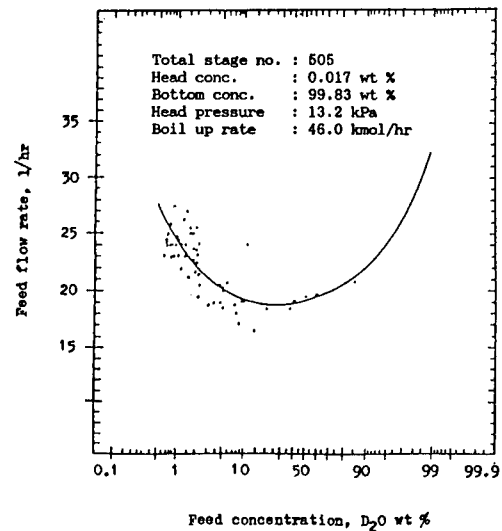


Fig. 8. Comparison of the Calculated Feed Flow Rates with Those of Actual Operation Data in the PHT Upgrader

one of 16 feed points, which is selected to match the concentration of the feed with that of vapour in the tower. With the assumption and 5 input variables listed in Table 2, the feed flow rate can be calculated as a function of concentration by the computer program.

In Fig. 8, the calculated feed flow rates are compared with those of actual operation data in the PHT upgrader. The program outputs are in good agreement with the actual data, although the data points are randomly obtained from the operation of the Wolsung upgrader. Therefore, it may be concluded that the computer program can predict output variables properly.

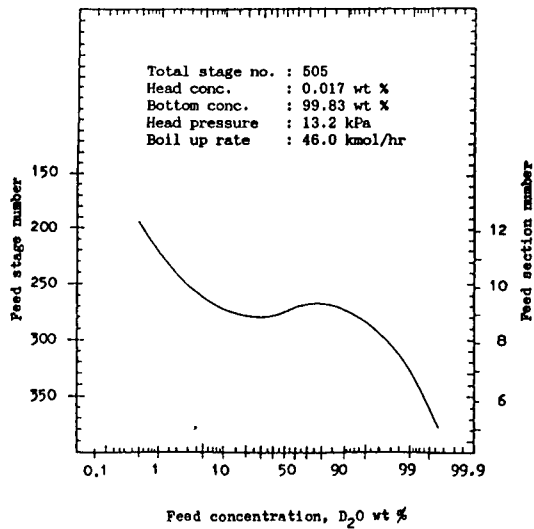


Fig. 9. Feed Stage and Section Number as a Function of Feed Concentration in the PHT Upgrader

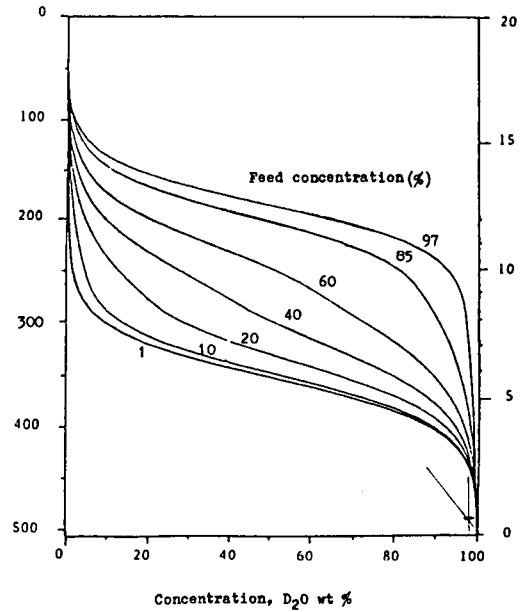


Fig. 10. Concentration Profiles as a Function of Feed Concentration in the PHT Upgrader

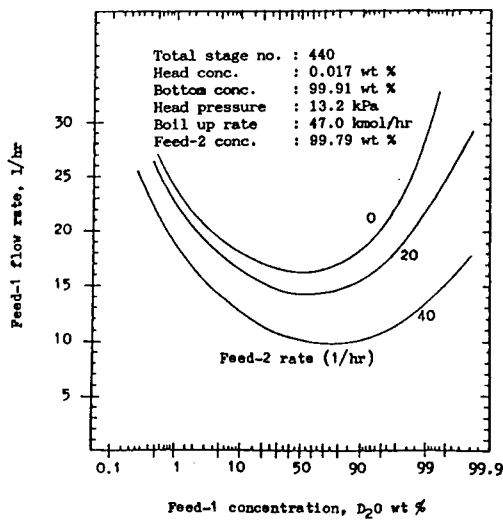


Fig. 11. Feed-1 Flow Rate as a Function of Its Concentration for Different Feed-2 Flow Rates in the Mod Upgrader

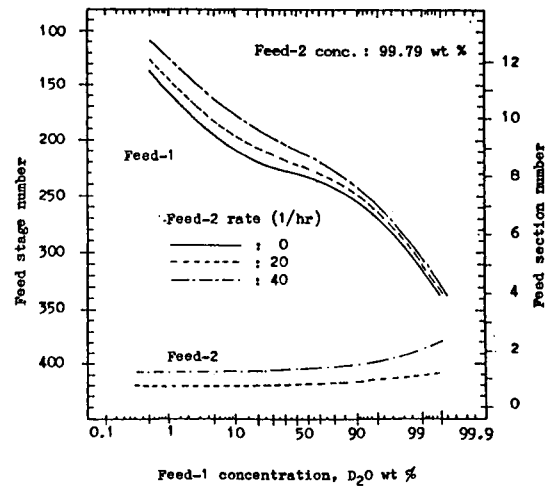


Fig. 12. Stage and Section Numbers of Feed-1 and Feed-2 as a Function of Feed-1 Concentration in the Mod Upgrader

IV-2. Application to Wolsung upgraders

The computer code developed in this study offers an operating guide for determination of flow rate and sec-

tion (or stage) number of feed, while maintaining the desired concentrations of head and bottom product. It is shown in Fig. 8 that the feed flow rate of the PHT upgrader has a minimum value at 25-30 wt % of feed concentration. On the other hand, feed stage number

of the PHT upgrader does not decrease monotonously with increasing feed concentration as shown in Fig. 9. For optimum operation of the upgrader, feed must be entered at section number 9 corresponding to stage number 265-290, when its concentration is in the range of 4-95 wt %. It is clearly illustrated in Fig. 10 that there is a wide difference of the concentration profiles in the PHT upgrader for different feed concentrations.

The Mod upgrader is designed to feed dually. One (feed-1) has the lower D₂O concentration from the collection system of the D₂O leakage, the other (feed-2) has the higher D₂O concentration from the main moderator system. The variation of feed-1 flow rate with its concentration is shown in Fig. 11 for three different feed-2 flow rates of D₂O concentration of 99.79 wt %. As the feed-2 flow rate increases, feed-1 flow rate decreases and the feed-1 concentration at the minimum value of its flow rate tends to increase. Stage numbers of feed-1 and feed-2 with feed-1 concentration as the parameter for different flow rates are shown in Fig. 12. The figure shows that feed-1 stage number increases monotonously with increasing concentration, but feed-2 stage number remains almost invariable.

Also, computer code can be used for the determination of the optimum conditions, such as determining the desired concentration of head product. The amount of the loss in operating an upgrader can be estimated by operating cost of the upgrader and the loss of D₂O in the head product to be released. Thus, the amount of the loss per kg of feed, COST, can be defined as follows.

$$\text{COST} = (\text{Dloss} \cdot \text{C1} + \text{C2}) / \text{Feed}$$

where Dloss: flow rate of D₂O in head product (kg/hr)

C1 : a cost of D₂O (186, 168 Won/kg)

C2 : operating cost of steam, electric energy, cooling and chilled water (6,178 Won/hr)

Feed : feed flow rate (kg/hr)

As the D₂O concentration of head product increases, feed flow rate (Feed) and D₂O loss (Dloss) increase simultaneously. However, the operating cost is almost

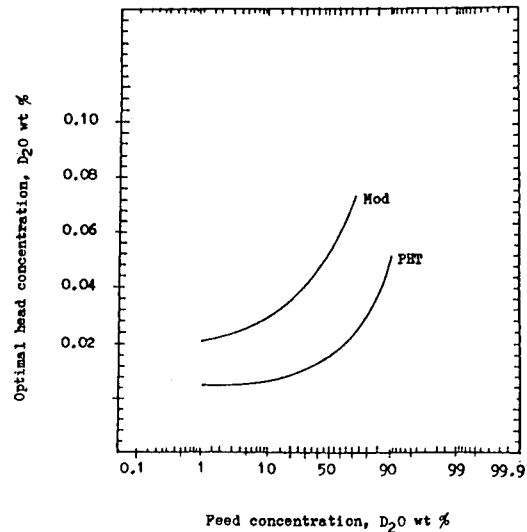


Fig. 13. Optimum Head Concentrations as a Function of Feed Concentration in the Wolsung D₂O Upgraders

constant. Therefore, the optimum head concentration exists at the operating condition where the amount of loss is minimized. Fig. 13 shows the optimum head concentrations as a function of feed concentration in the two upgraders. The optimum concentration increases with feed concentration. Also, the optimum concentration of the PHT system is higher than that of Mod system due to the difference of the number of total stages of the two systems.

Head concentrations have been specified to 0.017 wt % of D₂O in both upgraders, before this computer code was developed. The upgrader loss at the Wolsung plant can be reduced by using the newly optimized guide obtained by the computer code.

V. Conclusion

A computer code has been developed for the process analysis and the design of the heavy water upgraders in this study. It may be concluded that this code is suitable for the process analysis of Wolsung D₂O upgraders, since its calculated outputs agree well with the actual upgrader data. The computer code can

offer operating guides such as control of flow rate and selection of stage number of feed to obtain specified output concentrations of the products. Knowledge of the optimum concentration of head product calculated by the computer code can also be used to cut down the operating expenses of the upgraders. Thus, the computer code is now applied to the upgraders at Wolsung nuclear power plant for its efficient operation.

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