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HYDROGEN SEPARATION FROM THE H_2/N_2 MIXTURE BY USING A SINGLE AND MULTI-STAGE INORGANIC MEMBRANE

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Abstract–The separation characteristics of hydrogen from a gas mixture were investigated by using a single and two-stage inorganic membrane. Three palladium impregnated membranes were prepared by using the sol-gel, hydrolysis, and soaking-and-vapor deposition (SVD) techniques. A two-stage gas separation system without a recycling stream was constructed to see how much the hydrogen separation factor would be increased. Numerical simulation for the separation system was conducted to predict the separation behavior for the multi-stage separation system and to determine the optimal operating conditions at which the highest separation factor is obtained. Gas separation through each prepared membrane was achieved mainly by Knudsen diffusion. The real separation factor for the H_2/N_2 mixture was increased with the pressure difference and temperature for a single stage, respectively. For the two-stage separation system, there was a maximum point at which the highest separation factor was obtained and the real hydrogen separation factor for H_2/N_2 mixture was increased about 40% compared with a single stage separation. The numerical simulation for the single and two-stage separation system was in a good agreement with the experimental results. By numerical simulation for the three-stage separation system, which has a recycle stream and three membranes that have the same permeability and hydrogen selectivity near to the Knudsen value, it is clear that the hydrogen separation factors for H_2/N_2 mixture are increased from 1.8 to 3.65 and hydrogen can be concentrated up to about 80%. The separation factors increased with increasing recycle ratio. Optimal operating conditions exist at which the maximum real separation factor for the gas mixture can be obtained for three-stage gas separation and they can be predicted successfully by numerical simulation.

Key words : Inorganic Membrane, Hydrogen Separation, Multi-Stage Membrane

INTRODUCTION

The inorganic membrane has been studied for gas separation due to its thermal and chemical stability, and membrane gas separation technology is accepted in the chemical and petroleum refining industries. Among the many types of gas separation, there has been interest in the removal of hydrogen from purge gases, the separation of H_2/CO in synthesis gases, oxygen and nitrogen separation from air, dehumidification of gas streams and the recovery of high purity hydrogen [Tsuru et al., 1995; Bhide et al., 1991].

Compared to polymer membranes, porous inorganic membranes have suffered from low selectivity. In general, a more selective membrane inevitably has less permeability. Many efforts have been made to overcome such disadvantages [Uhlhorn et al., 1992; Ha et al., 1996; Lee et al., 1994]. The separation factor obtained by the Knudsen diffusion of gases through a micro-porous inorganic membrane cannot be over the Knudsen limit, which is the square root of the mass ratio of each gas component. In order to overcome this Knudsen limit, the surface diffusion is introduced by modifying the surface or pores of the inorganic membrane [Lee et al., 1995; Yoon et al., 1996]. However, this research has focused on the preparation of the selective membrane. Considering the application of the inorganic membrane technique for gas separation from coal gasification gas or the synthesis gas mixture, it is necessary to do research on the separation characteristics of hydrogen from the gas mixture.

There have been other engineering approaches to enhancing the separation performance of membranes. It is the design of membrane modules and their optimal configuration that leads to a higher separation factor [Hwang et al., 1980; McCandless et al., 1985; Qiu et al., 1991; Stern et al., 1984]. It was reported that the recycle of the permeate or retentate stream to the inlet stream can increase the separability of the used membrane [Qiu et al., 1991]. In a multi-stage membrane separation, the configuration of membranes is a critical factor for obtaining the desired gas with a high purity from the gas mixture. Research about numerical simulation to a multi-stage separation system is required to predict the effect of system variables such as recycle ratio, intermediate pressure, and recycling structure on the membrane performance because it is difficult to prepare membranes with the same performance and to test the effect of system variables directly. Although the optimal conditions for a multi-stage system vary with the properties of the membrane used, further research for multi-stage separation is required to predict the optimal condition.

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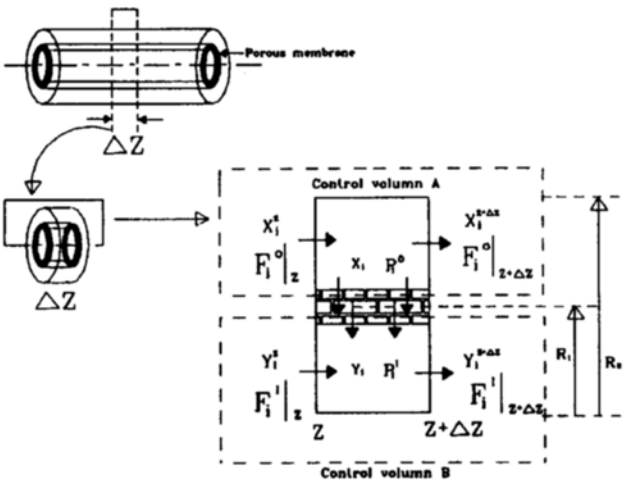

Fig. 1. Schematic diagram of the control volume for deriving the model equations.

The purpose of this work is to understand hydrogen gas separation from a gas mixture, such as H_2/N_2 by the porous inorganic membrane where the gas separation is achieved mainly by Knudsen diffusion, and to provide information for the preparation and selection of a porous membrane suitable for obtaining the desired gas with high purity from a gas mixture. Three membranes were prepared by the sol-gel, hydrolysis, and soaking-and-vapor deposition (SVD) techniques [Lee et al., 1995; Yoon et al., 1996]. The gas separation mechanism for the prepared membrane was investigated by the measurement of pure gas permeability as a function of pressure difference from 25 to 400 °C. A two-stage gas separation system was constructed to investigate the separation characteristics and see how much the hydrogen separation factor would be increased. A three-stage separation system, which is expected to increase the performance of membranes, was proposed, and the characteristics of hydrogen gas separation from H_2/N_2 were investigated by numerical simulation.

MODELING

Fig. 1 is a schematic diagram showing the membrane separation system to derive the model equations. The mass balance for the retentate and permeate side can be established at the infinitesimal control volume shown in Fig. 1. The control volume A and B represent the retentate and permeate side, respectively. The mass balances were applied to the control volume A and B under the following assumptions :

1. The system is at steady state.
2. The flows in the retentate and permeate stream are plug flow.
3. All components in the feed are permeable through the membrane.
4. The permeability of each gas component is the same as that of the pure gas, and is a constant along the membrane at a fixed pressure.
5. The pressure drop both for the retentate and permeate stream is negligible. So the total pressure of the gas phase is constant along the membrane.

Applying the mass balance to component i for the control volume A and B, the governing equations were obtained as follows.

$$\frac{dXi}{d\xi}=-D_i\left(a\frac{X_i}{\Sigma X_i}-\frac{Y_i}{\Sigma Y_i}\right) \qquad (1)$$

$$\frac{dXi}{d\xi}=D_i\left(a\frac{X_i}{\Sigma X_i}-\frac{Y_i}{\Sigma Y_i}\right) \qquad (2)$$

where, $D_i=2\pi R_{log-mean}\alpha_i\frac{L}{F_T}P^l$

$$\sum_i\frac{d(X_i+Y_i)}{d\xi}=0 \qquad (3)$$

B.C : At $\xi=0$; $X_i=X_{i,o}$, $Y_i=Y_{i,o}$

All notations used in this equation are listed in the nomenclature section.

Fig. 2 is a schematic diagram for the three-stage separation system with recycle stream. All permeates of the first stage flow to the retentate side of the second stage. A part of the retentate stream of the second stage is recycled to the permeate side of the first stage. The permeate stream from the second stage flows to the retentate side of the third stage and some retentate of the second stage is recycled into the permeate side of the second stage.

The R_1 and R_2 are defined as the recycle ratio of the retentate from the second stage to the first stage and from the third stage to the second stage, respectively. The pressure at the permeate side of the first stage should be the same as that of P_2 at the retentate side of the second stage as shown in the Fig. 2. Also, the pressure at the permeate side of the second stage is the same as that of P_3 at the retentate side of the third stage.

The governing equations for each stage correspond to Eqs. (1) and (2). Therefore, model equations used in a single stage can be applied to the three-stage simulation. The iteration scheme for the multi-stage is the following :

1. Guess the inlet fraction, $Y_{i,0}^1$, $Y_{i,0}^2$ of the permeate side for each stage.
2. For the first stage, predict the values of X_i^1, Y_i^1 by the simulation of a single stage.
3. Take Y_i^1 as the input values for the second stage :

$X_{i,0}^2=Y_i^1$

For the second stage, repeat step 2 to predict the value of X_i^2, Y_i^2.

4. Take Y_i^2 as the values of $X_{i,0}^3$.

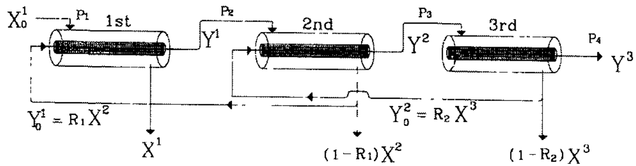

Fig. 2. Schematic diagram of the three-stage gas separation.

For the third stage, repeat step 2 to predict the value of X_i^3, Y_i^3.

5. Compare the predicted values with the guessed value in the step 1.

Thus, $Y_{i,0}^1$ with $(R_1 \times X_i^2)$

Y_{i0}^2 with $(R_2 \times X_i^3)$

Then, calculate the error between the guessed value and the predicted value.

6. If the error is not in the error boundary, then take $(R_1 \times X_i^2)$ to a new and $(R_2 \times X_i^3)$ to $Y_{i,0}^2$.

7. Repeat the steps from 1 to 6 when the error goes into the error boundary.

In general, the real hydrogen separation factor is defined by

$$\alpha = \frac{y/(1-y)}{x/(1-x)} \tag{4}$$

where y and x is the hydrogen composition at the permeate and the feed stream, respectively.

EXPERIMENTS

1. Preparation of the Inorganic Membrane

In this section, the preparation methods of each membrane will be described briefly. The detailed procedure is described elsewhere [So et al., 1997; Yoon et al., 1995]. The sol-gel process, in-situ hydrolysis, in-situ silica sol coating and soaking vapor deposition (SVD) techniques were applied to modify the pore of three inorganic membranes. The porous support used in this work is an a-alumina tube (Noridake, Co) which has a length of 500 mm, outer diameter of 10 mm, and inner diameter of 7 mm. The porous membrane used in this work has an asymmetric structure and an average pore size of 80 nm in the dense layer.

1-1. Sol-Gel Step

First, the pore of the membrane was modified by the sol-gel process. The preparation of alumina sol followed the method developed by Yoldas [Yoldas et al., 1973]. Aluminum iso-proposide ($Al[CHO(CH_3)_2]_3$, Aldrich Chemicals Co.) was used as an alkoxide precursor. For the impregnation of catalytic material, which can help the adsorption of the hydrogen on the pore surface and into the pore of the ready-made membrane, the palladium particles were impregnated directly into the alumna sol. The palladium chloride, $PdCl_2$ (SIGMA,Co.), was used as the precursor of the palladium particles. The prepared palladium sol was sonicated to disperse well the palladium particles in the sol solution. Thereafter, the bare porous membrane was dipped in the sol solution, dried at room temperature, and calcined at 500 °C. Finally, it was reduced at 400 °C.

1-2. Hydrolysis Step

The in-situ hydrolysis proceeded by using the TEOS (Tetra Ethyl Ortho Silicate, Adrich chemical Co.). TEOS and water was vaporized and each vapor was carried by carrier gas, which was nitrogen, into the outside and inside of the porous membrane, respectively.

1-3. Soaking and Vapor Deposition (SVD) Step

Table 1. Preparation history of each membrane

Membrane	Preparation history
A	sol-gel #7+in-situ hydrolysis #2+SVD #4+ESVD#2
B	sol-gel #8+in-situ hydrolysis #2+SVD #5+ESVD#2
C	in-situ sol-gel #6+SVD #6+silica-sol coating #6+ESVD #2

The SVD technique was applied to plug the pore of the ready-modified membrane and impregnate the palladium particles into its pore. The palladium(II) acetate, which was used as the palladium precursor, was dissolved in the acetone of 200 ml with the HCl of 5 g. This solution was supplied to the outside of the membrane that had been modified by the sol-gel and in-situ hydrolysis step. Then, the palladium acetate solution was diffused into the pore of the membrane by the capillary force. The inner pressure of the membrane was maintained under atmospheric pressure : for example, 100 to 150 torr for about 15 min to easily take the palladium solution near the surface pores to the inner pore of the membrane, before drying it to remove the solvent of acetone. Thereafter, the membrane was dried in the drying oven at 300 °C for 3 hours to form the precursor of the palladium acetate in the pore. Then, the membrane impregnated by the palladium acetate was heated at 180 °C to form the intermediate layer of the palladium in the porous support. This process is called enhanced soaking and vapor deposition (ESVD) [Lee et al., 1995].

Three membranes were prepared by the above techniques and have the preparation history as shown in Table 1. The # number means the repeating times of each step, Therefore, for example, the membrane A was prepared by 7 times of sol-gel, 2 times of in-situ hydrolysis, 4 times of SVD, and 2 times of the ESVD step. The ESVD step is the same as the SVD technique except for applying the vacuum into the inner side of the membrane during the ESVD.

2. Measurement of the Permeability for Pure Gases

The permeability and hydrogen selectivity of pure gases such as H_2, and N_2 were measured as a function of the pressure difference which ranges between 170.3 and 411.7 kPa. Fig. 3(A) is the schematic diagram for measuring the permeability of pure gases. The pressure at the retentate side was controlled by a needle valve installed in the retentate stream as shown in Fig. 3(A). The inlet flow rate of each gases was controlled by the Mass flow controller (MFC, UFC-1500A, Unit instrument Inc.) at 2 *l*/min.

3. Separation of Hydrogen from the Gas Mixture

3-1. Single Stage

Hydrogen separation from the gas mixture was conducted for the prepared membranes. The flow rates and compositions of each of the gases for the H_2/N_2 mixture were controlled by the mass flow controller (MFC, UFC-1500A, Unit instrument Inc.). The hydrogen composition at the inlet stream of the H_2/N_2 was maintained at 50 vol%. The permeate stream was kept to atmospheric pressure. The pressure of the retentate was changed from 170 kPa (1.68 atm) to 412 kPa (4.06 atm) by using the needle valve which is connected with the

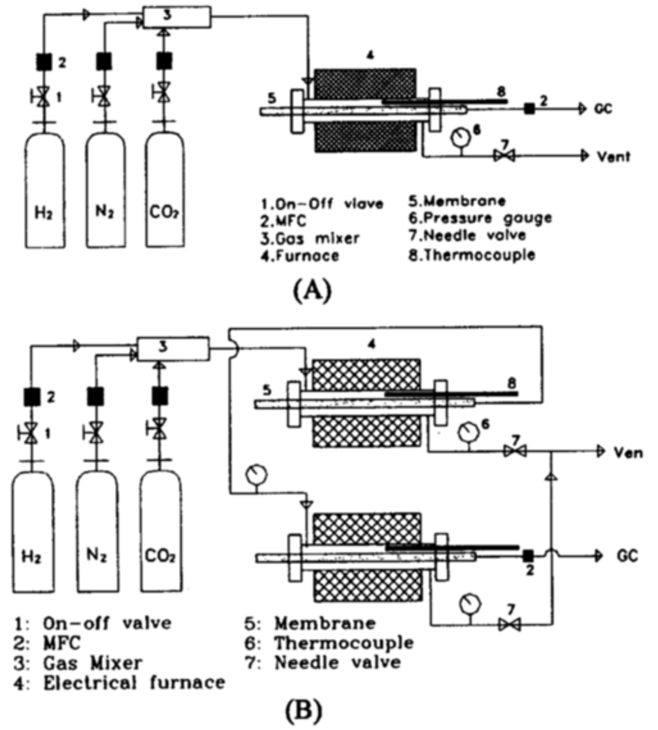

Fig. 3. Apparatus for gas separation.
(A) single, (B) multi-stage

retentate stream. The module was maintained between 25 to 400 °C by using an electrical furnace with a temperature controller.

3-2. Two-Stage

The experimental apparatus for the two-stage separation is shown in Fig. 3(B). The first stage was set with the membrane C which had much higher permeability than that of membrane B which was set on the second stage. All permeates from the first stage were made to flow into the retentate side of the second stage. The pressure, P_1 at the retentate side of the first stage was changed between 239.3 to 411.7 kPa by the needle valve connected with the retentate stream of the first stage. The permeate flowing into the retentate side of the second stage was controlled by the needle valve to produce the proper pressure, P_2 at the retentate side of the second stage. No pump was used in order to make the permeate of the first stage flow to the retentate side of the second stage. Thus, the pressure at the permeate side of the first stage should be same as the one at the retentate side of the second stage.

The pressure at the retentate side of the first stage was maintained at a fixed value when the pressure at the retentate side of the second stage was changed. Therefore, the flow leaving from the retentate side of the first stage was adjusted to keep the pressure at the retentate side of the first stage at a fixed value.

4. Analysis of the Permeate Composition for the Gas Mixture

The gas composition of the permeate for each gas mixture was analyzed by gas chromatography (GC, HP 5890 Series II) with a thermal conductivity detector (TCD) and porapark Q column. A part of the permeate for each gas mixture flowed to the GC with an autosampling valve.

RESULTS AND DISCUSSION

1. Experiment

Fig. 4 shows the permeability of H_2 and N_2 for the prepared membranes as a function of the pressure difference, P^R-P^P, at 25 and 400 °C. From this figure, it is clear that the gas flow mechanism through pores of the prepared membranes is achieved mainly by the Knudsen diffusion.

1-1. Single Stage

The ideal hydrogen selectivity to the nitrogen for membrane A was plotted as a function of pressure difference at several temperatures as shown in Fig. 5. At low pressure difference and a temperature of 400 °C, the hydrogen selectivity was above the Knudsen limit (=3.74). However, the ideal hydrogen separation factor had a lower value than the Knudsen limit at the high pressure regime because there were the relatively large pores in the modified membrane and Poiseuille flow through the large pores occurred. The contribution of Poiseuille flow to gas permeation was increased with increasing the pressure difference. The dependency of the real hydrogen separation factor for the H_2/N_2 mixture on the pressure difference and the temperature is shown in Fig. 6. The real

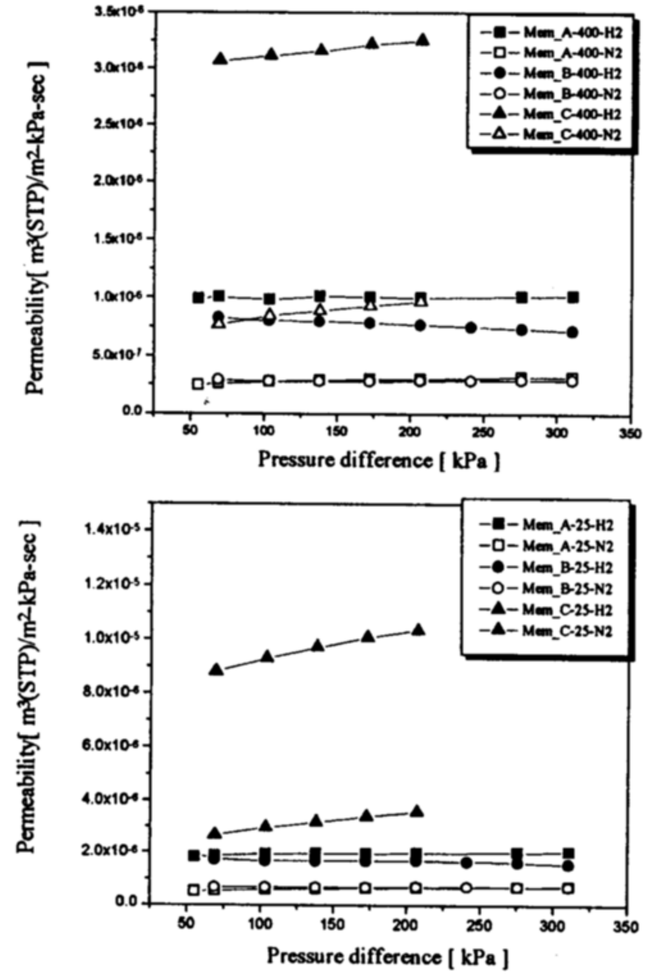

Fig. 4. Permeability of pure gases as a function of pressure difference.

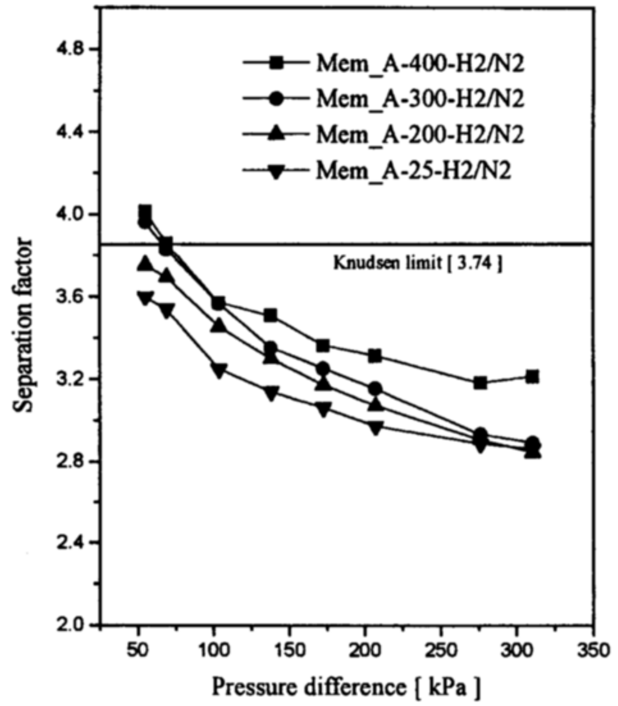

Fig. 5. Ideal separation factor of hydrogen to nitrogen as a function of pressure difference between 25 and 400 °C for membrane A.

hydrogen separation factor was increased with increasing the pressure difference and the temperature.

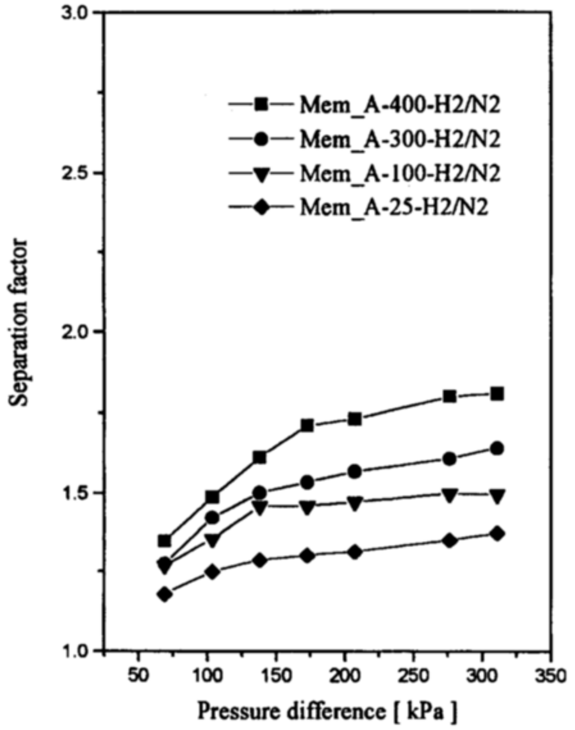

Fig. 6. Separation factor of hydrogen to nitrogen as a function of pressure difference at several temperatures for membrane A.

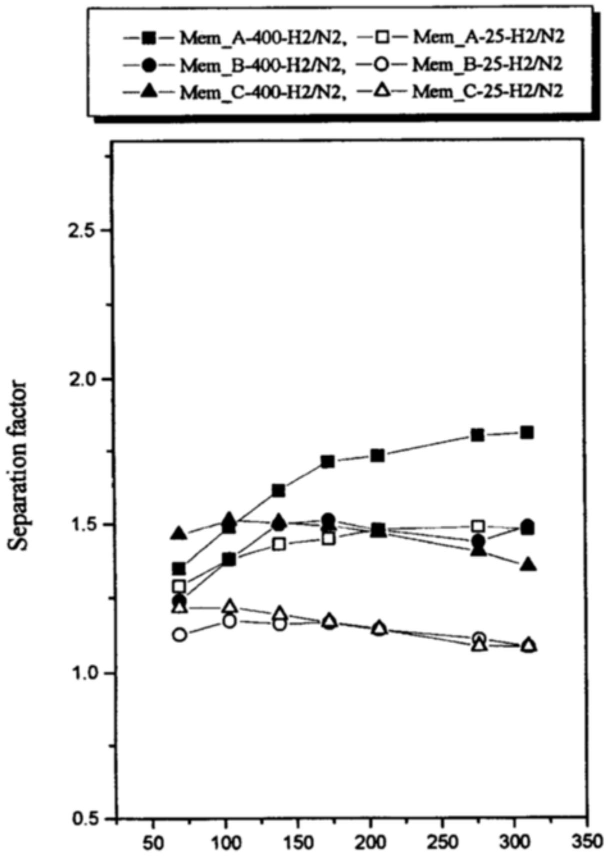

Fig. 7. The real hydrogen separation factors for membrane A, B and C at 25 and 400 °C.

In general, the real separation factor is smaller than the ideal separation factor due to backdiffusion, non-separative diffusion, concentration polarization on the feed or permeate side and the occurrence of Poiseuille flow in large pores [Yan et al., 1994]. So, the increase of the pressure difference reduces the backdiffusion, which results in increasing the real separation factor. The maximum separation factor of hydrogen to nitrogen obtainable by membrane A was about 1.8 at a pressure difference of 310 kPa and 400 °C. The real hydrogen separation factor for all prepared membranes is plotted in Fig. 7 which shows that the separation factor of hydrogen for the H_2/N_2 mixture is increased with the pressure difference and temperature.

1-2. Two-Stage

The real hydrogen separation factors for two-stage system are plotted in Fig. 8(a) as a function of intermediate pressure at a fixed total pressure. The separation factor showed a maximum point with changing pressure of the second stage under a fixed pressure of the retentate side of the first stage. The maximum separation factor at each fixed P_1 is replotted in Fig. 8(b) as a function of the total pressure difference. The obtainable maximum hydrogen separation factor, which was about 2.1, was achieved experimentally at P_1 of 308.1, P_2 of 203.3 kPa and 400 °C. The real separation factor by the two-stage system was increased about 40 % compared with the single stage in which the maximum hydrogen separation factor to nitrogen was about 1.5 for both membranes B and C.

2. Numerical Simulation

The numerical simulation for a single stage separation was

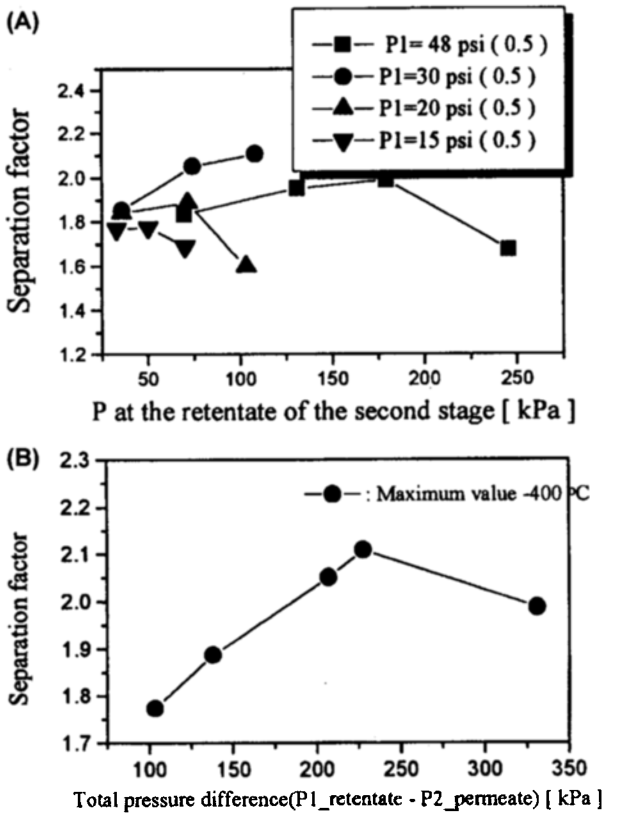

Fig. 8. (A) The real hydrogen separation factor as a function of the intermediate pressure, P_2, (B) The maximum separation factor at a fixed P_1 for two-stage separation system.

conducted by using the pure gas permeability as a function of pressure difference. Although the gas flow through pores of the prepared membrane was achieved mainly by the Knudsen diffusion, the Poiseuille flow affected the gas separation with increasing high pressure difference. So, the permeability of pure gases can be written as a function of the pressure difference.

$$\alpha_i = K_i + G_i \Delta P$$

Fig. 9 shows the simulation results of the H_2/N_2 mixture for the prepared membrane A. The simulation results were in good agreement with the experimental results. The results of the numerical simulation for the two-stage system are shown in Fig. 10. The thin solid line in Fig. 10 indicates the results obtained by using the permeability of each pure gas for the two membranes used. The simulation results showed somewhat of a deviation from the experimental results, although the trend of the simulated separation factor was consistent with that of the experiment. Jung et al. reported that the porous inorganic membrane by the pore modification techniques used in this work showed a decrease of separation performance with the usage time [Jung et al., 1997]. It was supposed that the performance of each used membrane in the two-stage would be decreased when the two-stage experiment was conducted. Therefore, the value of K in the above equation of pure gas permeability for any membrane was increased to lead the numerical expectation to the experimental value. When the per-

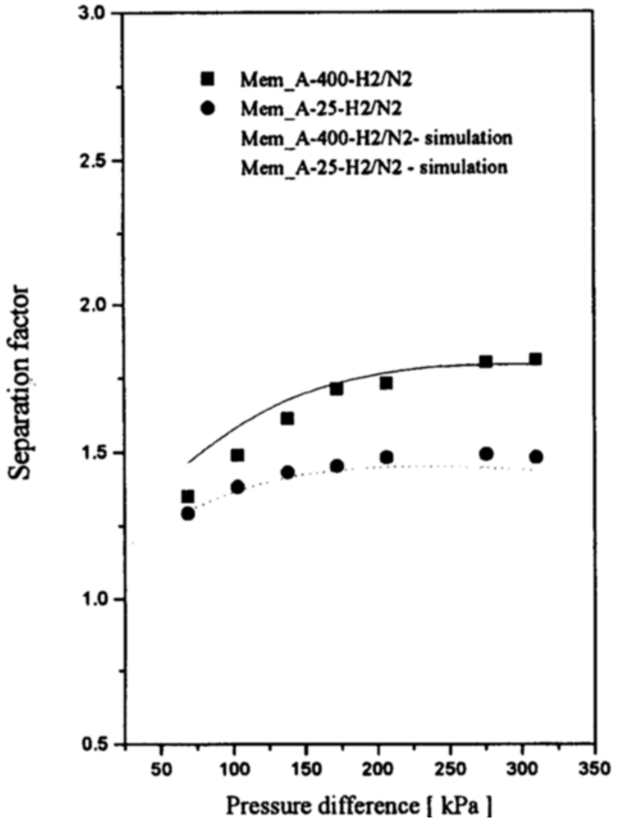

Fig. 9. Simulation and experimental results for the hydrogen separation from H_2/N_2 mixture for the membrane A.

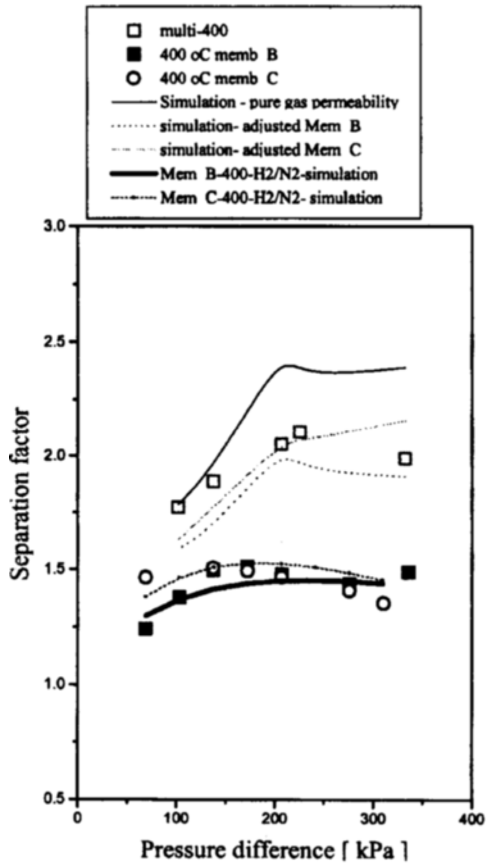

Fig. 10. The simulation results for two-stage separation.

meability of each gas is adjusted for the membrane B or C, the simulation results are in a good agreement with the experimental results.

Three-stage simulation results are shown in Figs. 11 to 13. The pressure at the retentate of the first stage was fixed at 405.3 kPa (=4 atm). The permeate side of the third stage was assumed to be open to the atmosphere, thus has a fixed pressure of 101.3 kPa (=1 atm). Therefore, the total pressure difference of the three-stage gas separation system is the same as that for a single stage system. The pressures of each stage are given by P_i , where P_1 denotes the pressure at the retentate side of the first stage, P_2 is the pressure at the permeate side of the first stage or at the retentate side of the second stage, P_3 is the pressure at the permeate side of the second stage or at the retentate side of the third stage, and P_4 is the pressure at the permeate side of the third stage.

The effect of the intermediate pressure such as P_2 and P_3 was investigated at a fixed recycle ratio, R_1 and R_2 which are represented from the second stage to the first stage and from the third stage to the first stage. Fig. 11 shows the real hydrogen separation factor as a function of P_2 and P_3. The hydrogen separation factors were increased with decreasing P_3 at a fixed P_2 and reached a maximum value with changing P_2 at a fixed P_3. The decrease of P_2 value at a fixed P_3 means that the driving force of the first stage and the second stage is increased and decreased, respectively. The increase of the P_3 value at a fixed P_2 means that the driving force of the second stage and the third stage is decreased and increased, respectively. Under the fixed recycle ratio as a value of 0.5, the maximum hydrogen separation factor to nitrogen can be obtained at P_2 of 187.5 kPa (=1.85 atm) and P_3 of 121.6 kPa (=1.2 atm).

The effect of the recycle ratio, R_1 and R_2, on the real separation factor is showed in Fig. 12. The real separation factor of hydrogen to nitrogen was increased with increasing recycle ratio R_1 and R_2. But, at an R_1 and R_2 value over 0.9, the separation factors decreased slightly. The maximum separation factors obtainable under the fixed R_1 and R_2 as the value of 0.9 are shown in Fig. 13 as a function of P_2 and P_3. The real hydrogen separation factor showed the maximum point with changing the P_2 and P_3. The maximum separation factor of hydrogen to nitrogen achievable by the three-stage separation system with recycle stream was about 3.65 at a P_2 of 268.5 and a P_3 of 131.7 kPa under the R_1 and R_2 of 0.9, respectively. This separation factor is much greater than that of the single stage separation under the same driving force. From the above result, it is clear that the proposed three-stage separation system promises to enhance the membrane performance to lead to a high real hydrogen separation factor up to 3.65, even though the ideal gas separation factor for the used membrane was less than the Knudsen value and the real hydrogen separation factor for a single stage of the membrane was about 1.8.

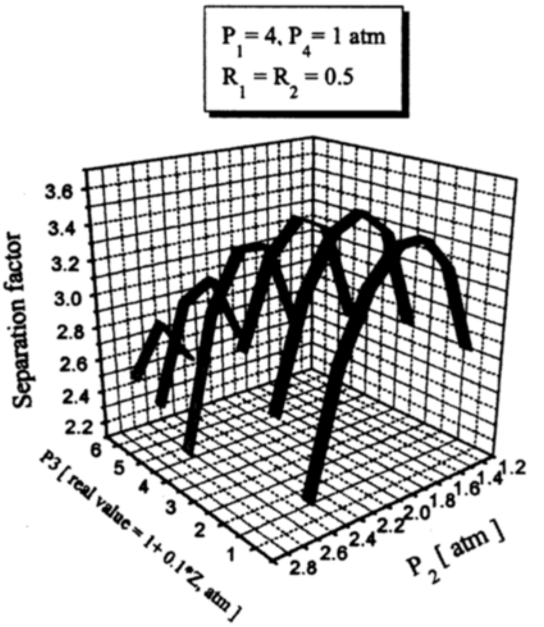

Fig. 11. Hydrogen separation factor for H_2/N_2 mixture as a function of P_2 and P_3 at the fixed recycle ratio of 0.5 for three-stage separation system.

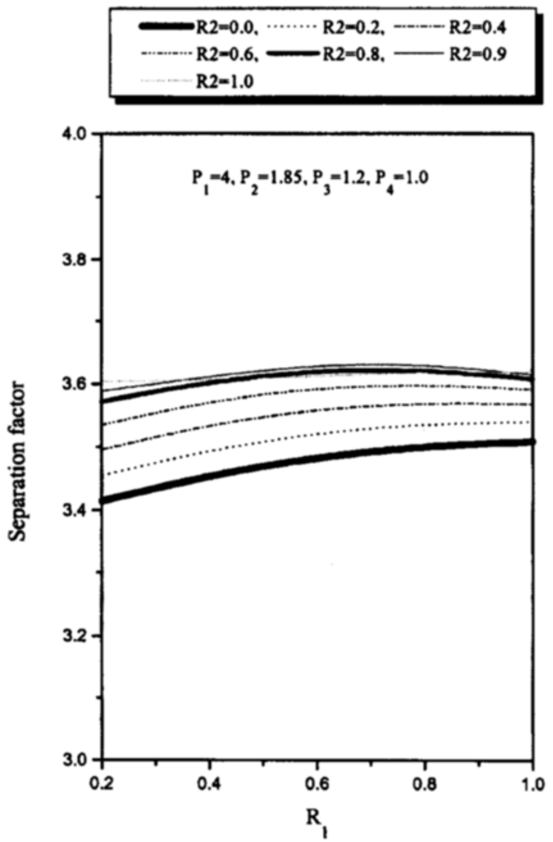

Fig. 12. Hydrogen separation factor with the recycle ratio at fixed pressures for three-stage separation system.

The maximum separation factors obtainable from the proposed three-stage membrane system were simulated with changing the total driving force, P_1-P_4. Table 2 shows the effect of the total driving force on the maximum separation factor as a function of P_2 and P_3. At a P_1 of 506.6 kPa (=5 atm), the maximum hydrogen separation factor to the nitrogen achievable by the three-stage separation system was over the Knud-

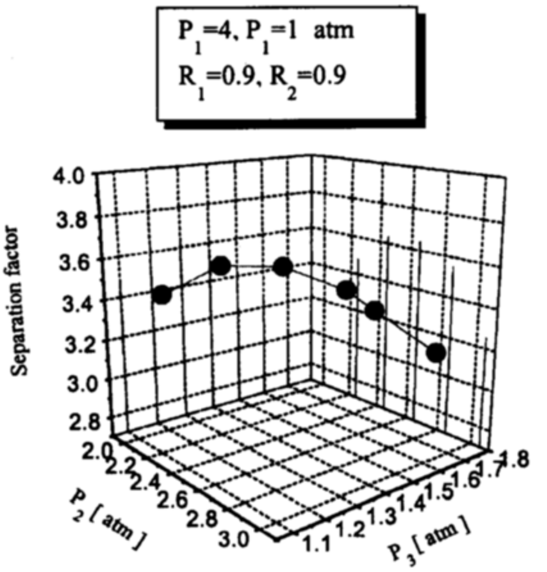

Fig. 13. Maximum separation factor of hydrogen for H_2/N_2 mixture at a given recycle ratio in the three-stage separation system.

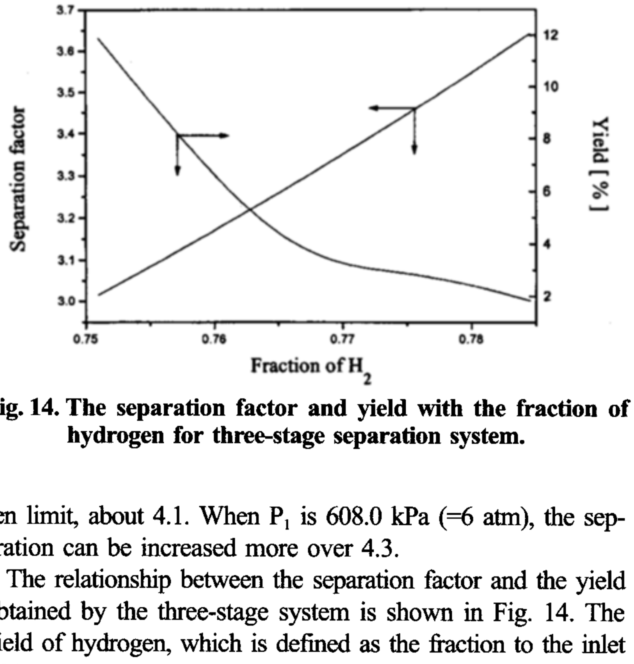

Fig. 14. The separation factor and yield with the fraction of hydrogen for three-stage separation system.

sen limit, about 4.1. When P_1 is 608.0 kPa (=6 atm), the separation can be increased more over 4.3.

The relationship between the separation factor and the yield obtained by the three-stage system is shown in Fig. 14. The yield of hydrogen, which is defined as the fraction to the inlet of hydrogen obtained at the permeate side of the final stage, decreased with the increase of the hydrogen separation factor.

CONCLUSIONS

The palladium-impregnated inorganic membranes were prepared by sol-gel, in-situ hydrolysis, and SVD techniques and the separation characteristics of hydrogen from the H_2/N_2 gas mixture were investigated.

The dominant gas transport mechanism through pores of the prepared porous inorganic membranes was the Knudsen diffusion. The ideal hydrogen separation factors for the prepared membranes were above the Knudsen limit due to the occurrence of surface flow at low pressure and high temperature. However, the considerable Poiseuille flow became dominant as the pressure difference was increased.

The real hydrogen separation factors for the prepared membrane were increased with increasing pressure difference and temperatures. The maximum separation factor to nitrogen achieved by the prepared membrane A was about 1.8 at a pressure difference of 310 kPa and 400 °C.

Two-stage gas separation system without a recycling stream was constructed. The real hydrogen separation factor to nitrogen was increased 40 % compared with the value of the single stage. There existed a optimum intermediate pressure at which the maximum separation factor was obtained.

Numerical simulation was conducted on both the single and three-stage membrane to see if the theoretical prediction of the real hydrogen separation factor, by using the permeability of pure gases, agreed with the experimental results, and how much the real separation factor of hydrogen could be increased by the proposed three-stage separation. The optimal operating conditions for the three-stage gas separation at which the real separation factor has a maximum value were predicted by the numerical simulation. The simulation results were in good agreement with the experimental results for a single stage and two-stage.

The hydrogen separation factor was increased from 1.8 to 3.6 by using the three-stage separation system for which membranes have the same permeability as the prepared membrane A. For the three-stage separation system, the real separation factors were increased with increasing the recycle ratio R_1 and R_2 up to 0.9, respectively. There existed the optimum P_2 and P_3 at which the actual separation factor had a maximum value. They were predicted successfully by the numerical simulation.

When the total driving force was increased from 405.3 to 608.0 kPa, the real hydrogen separation factor for H_2/N_2 mixture could be over the Knudsen limit, although the membrane A used for the three-stage simulation had much lower

Table 2. The optimal P_2 and P_3 value predicted by the simulation of the three-stage separation system with changing the total pressure difference under the fixed R_1 and R_2 of 0.9

P_1 (kPa)	P_2 (kPa)	P_3 (kPa)	P_4 (kPa)	Maximum real separation factor (single stage)	Maximum real separation factor (three-stage)	Hydrogen fraction
405.3	268.5	131.7	101.3	1.8[a]	3.65[b]	0.785
506.6	268.5	132.7	101.3	-	4.10[b]	0.804
608.0	288.8	141.9	101.3	-	4.38[b]	0.814

[a]Experimental result for the membrane A at 400 °C and the pressure difference of 310 kPa
[b]Simulation results

value than the Knudsen limit for a single stage separation.

NOMENCLATURE

a, P^O/P^I: pressure ratio between the permeate and retentate side
P^O : pressure at the retentate side [atm]
P^I : pressure at the permeate side [atm]
F_i^o : molar flow rate of component I at the retentate side [mol/sec]
F_i^I : molar flow rate of component I at the permeate side [mol/sec]
X_i : dimensionless molar flow rate at the retentate side
Y_i : dimensionless molar flow rate at the permeate side
L : length of membrane [m]
F_T : total inlet molar flow rate [mol/sec]
α_i : permeability of pure i-component [m^3 (STP)/m^2-kPa-sec]
x : dimensionless length

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