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Comparison of Gas Permeability and Selectivity Between Alumina Membrane and Vycor Glass at High Temperatures*

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ABSTRACT

In this study, gas permeability and selectivity of Vycor glass and alumina membrane were compared by using H_2, CO_2, CO, CH_4 and N_2 gases at the temperatures of 323-823 K before and afterapplying the silica coating process. H_2 permeability decreased with increasing the temperature before silica coating both in Vycor glass and alumina membrane. However, H_2 permeability increased with increasing the temperature, that was an indication of activated transport after silica coating both in Vycor glass and alumina membrane. Lower permeability values and higher selectivities were obtained in Vycor glass membrane than alumina membrane having higher permeability and lower selectivities.

Keywords: Membrane; Hydrogen; Permeability; Selectivity

1. Introduction

Membrane separation of gaseous mixtures has come to attract much attention from the viewpoint of energy-conservative recovery of gases, especially at high temperature, where most of ordinary organic membranes can not be applied because of their thermal instability. In contrast, inorganic membranes, which seem quite stable at high temperature and against most chemicals, will preferably be applied to gas separation processes as in [1].

The preparation and application of ceramic membranes has received much attention in the past few years as in [2]. Ceramic membranes are technically important in separation and filtration as well as in catalytic reactions, because of their high thermal and chemical stability, long life time and good defouling properties in comparison with polymeric membranes as in [3,4].

Hydrogen permeable and selective silica membranes have attracted much interest in the membrane gas separation field due to the importance of hydrogen as an industrial feedstock for the production of fuels and many chemicals as in [5, 6]. Silica membranes are attractive since they are chemically and thermally stable while offering high permeability and selectivity for hydrogen as in [7].

Silica fabricated by the sol-gel process is known to have high surface area and microporosity as in [8-10]. The sol-gel process, in this case, refers to the controlled hydrolysis and polymerization of tetraethylorthosilicate(TEOS) in a water-alcohol solution as in [11,12]. The solution goes through an irreversible sol-gel transition. The resulting gel can be dried to a rigid silica-like material.

2. Experimental

The alumina membrane changing the particle size distribution from 3 μm to 70 nm with the diameter of 21 mm and the thickness of 1 mm was purchased from Germany. The Vycor glass having the mean particle size of 40 Angstrom with the diameter of 21 mm and the thickness of 1 mm was purchased from USA. Gas permeability tests of Vycor glass and alumina membrane were performed by using H_2, CO_2, CO, N_2 and CH_4 gases at constant volume-variable pressure measurement system between 323 and 823 K and H_2 selectivites over other gases were determined. Then, alumina membrane was dipped into LUDOX colloidal silica solution at 2-3 s, subsequently, it was calcined in the oven up to the temperature of 1173 K with heating rate of 1 K/min. After two hours passed at the temperature of 1173 K, calcination was continued with cooling rate of 1 K/min. However, sol-gel process was applied to Vycor glass by refluxing a solution of TEOS, H_2O, C_2H_5OH and HNO_3 in the ratio of 1: 1: 26: 11.76 respectively at 353 K for two hours. A sample of solution was diluted with C_2H_5OH in the ratio of 1:18 and membrane was dipped for a few seconds. It was then dried at 393 K for three hours with heating rate of 1 K/min and calcined at 673 K for three hours with the same heating rate. The dipping was repeated a second time with the dilution being 1:180.

After silica coating processes of Vycor glass and alumina membrane were completed, they were exposed to gas permeation experiments with using H_2, CO_2, CO, N_2 and CH_4 gases among 323 and 823 K and H_2 selectivites over other gases were detected.

3. Results and Discussion

Gas permeability test of samples including Vycor glass and alumina membrane before and after silica coating was fulfilled by using constant volume-variable pressure measurement system for the temperatures around 323 and 823 K and H_2 selectivites over other gases were determined. Permeability coefficient of each gas was calculated from (1) and selectivity was also detected from (2).

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$$F = \frac{Q}{A * \Delta P} \quad (1)$$

Q: flow rate of gas (m^3/s)
A: membrane crossectional area (m^2)
ΔP: pressure difference (Pa)
F: permeability coefficient ($m^3/m^2.s.Pa$)

$$\alpha \equiv \frac{Ppermeate}{Pfeed} \quad (2)$$

□P: pressure
α : selectivity

Variation of H_2 permeability with the temperature for alumina membrane before and after silica coating was given in **Figures 1** and **2** respectively.

Variation of H_2 permeability with the temperature for Vycor glass before and after silica coating was observed in **Figures 3** and **4** respectively as well.

As it was observed from **Figures 1**, **2**, **3** and **4**, H_2 permeability decreased with increasing the temperature before silica coating both in Vycor glass and alumina membrane. However, H_2 permeability increased with increasing the temperature, which was a characteristic of activated transport as in [13] after silica coating both in Vycor glass and alumina membrane. Moreover, H_2 selectivities over other gases for both membranes were given in **Tables 1** and **2**.

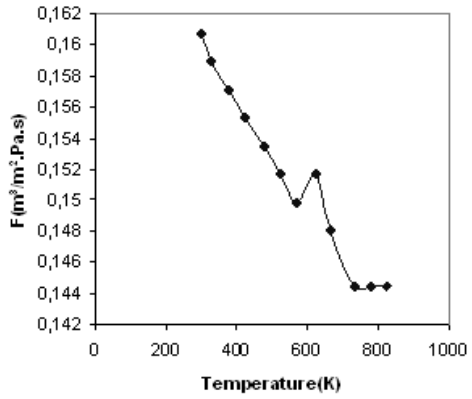

Figure 1. Variation of H2 permeability with temperature for alumina membrane before silica coating.

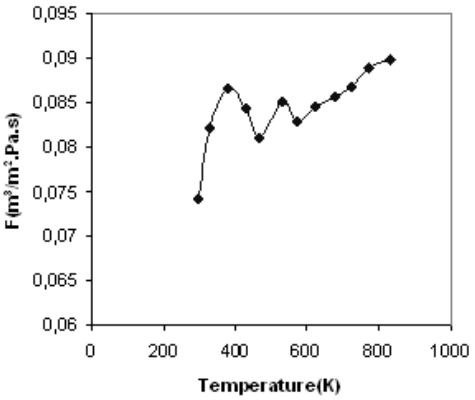

Figure 2. Variation of H2 permeability with temperature for alumina membrane after silica coating.

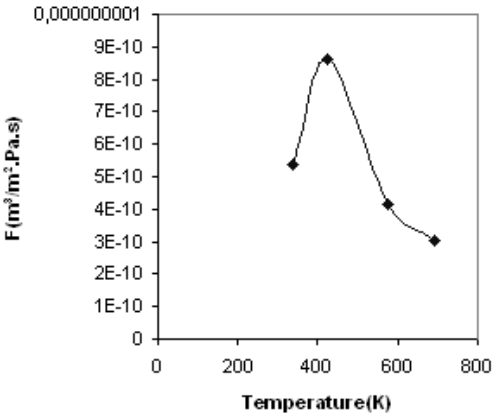

Figure 3. Variation of H2 permeability with temperature for Vycor glass before silica coating.

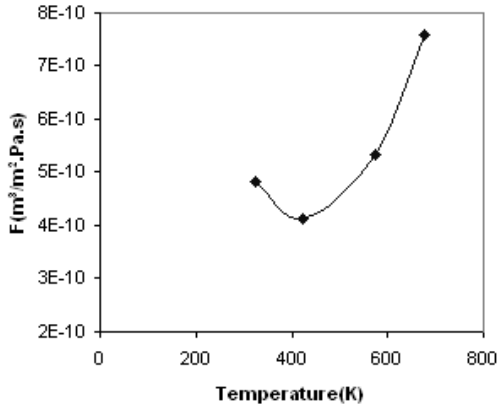

Figure 4. Variation of H2 permeability with temperature for Vycor glass after silica coating.

Table 1. H_2 selectivities over other gases for alumina membrane.

Alumina membrane		
Selectivities	Before silica coating	After silica coating
H_2/N_2	0,221118012	0,030482257
H_2/CO_2	0,148472744	1,777078454
H_2/CH_4	0,15335213	0,030238693

Table 2. H_2 selectivities over other gases for Vycor glass.

Vycor glass		
Selectivities	Before silica coating	After silica coating
H_2/N_2	6.515	4.712
H_2/CO_2	5.346	5.763
H_2/CH_4	4.475	10.468

According to the **Tables 1** and **2**, H_2 selectivities over N_2 and CH_4 gases decreased, while H_2 selectivity with respect to CO_2 gas increasing after silica coating in alumina membrane. This result may be due to the surface diffusion of gas molecules. However, H_2 selectivities over CO_2 and CH_4 gases increased, when H_2 selectivity in view of CO_2 gas decreased after silica coating in Vycor glass.

Results showed that alumina membrane with having higher permeability and lower selectivity, Vycor glass membrane having lower permeability and higher selectivity were obtained.

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