Preparation of Pore Filling Membrane with Pd Nanoparticles

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Abstract

Hydrogen perm-selective membranes were composed Pd nanoparticles. The nanoparticles were prepared by sonochemical reduction from Pd^{II} ions. Then the nanoparticles deposited on a substrate disc with electrophoresis technique. These electrophoretic membranes have shown high performance of perm-selectivity for H_2 with separation factor α=3.85, under room temperature.

Introduction

To realize the H_2 permeable membrane with high permeance rate, thinner metallic membrane has to be required[1)]. Thinner and denser membranes are required because of higher permeance rate of H_2.

In this work, Pd nanoparticles were prepared with sonochemical reduction technique. The nanoparticles were precipitated on anodic oxidation alumina disc, and then the perm-selective membranes were created successfully using electrophoresis technique. Permeances of H_2 were measured with a closed circulation system at room temperature.

Experimental

Pd nanoparticles were prepared from Pd^{II} solution (0.4mM) with ultrasonic reduction under Ar atmosphere for 20 min[2)]. Formation of nanoparticles was confirmed by UV-vis spectra. Morphology of nanoparticles was observed by high-resolution transmission electron microscope (HR-TEM). The prepared Pd nanoparticles were deposited on the surface of anodic aluminum oxide (AAO, pore size : 20nm) substrates by electrophoresis (electrophoresis condition : 500V, 3min). The morphologies of the deposited membrane were observed by field emission scanning electron microscope (FE-SEM).

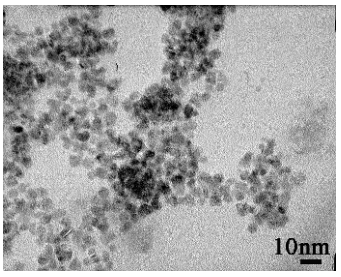


Fig.1 TEM photograph of Pd nanoparticles. (average particle size = 4.04 ± 1.28 nm)

Permiations of N_2 and H_2 were evaluated by monitoring the pressure on both sides of membrane using a vacuum line under ambient temperature.

Volume of permeation (q [cm^3]), permeability coefficient (R [cm^3 (cm^2 · mmHg · $sec)^{-1}$]), and separation factor (α) were calculated by equations described as follows,

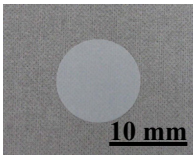
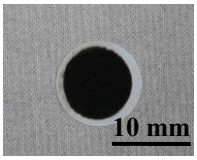
$$q = R(p_1-p_2)A\Delta t,$$

$$\alpha = R_{H_2} / R_{N_2},$$

where P is transmission coefficient [cm^3·cm (cm^2·cmHg·$sec)^{-1}$], (p_1-p_2) is differential pressure [cmHg], A is membrane area [cm^2], t is differential time [sec], l [cm] is membrane thickness.

Results and Discussions

Table 1. Gas permeation properties of obtained membrane

Membrane		Permeation gas	R [cm^3 (cm^2 ·cmHg ·$sec)^{-1}$]	α [-]
Before electrophoresis		H_2	1.97×10^{-6}	2.90
		N_2	6.77×10^{-7}	
After electrophoresis		H_2	9.37×10^{-8}	3.85
		N_2	2.43×10^{-8}	

From Fig. 1, average size of Pd nanoparticles was ca. 4.0 nm in diameter. Table 1 shows photographs of AAO substrates before and after electrophoresis of Pd nanoparticles. It was clear that Pd nanoparticles were fixed on the surface of substrate because AAO substrate colored to black after the electrophoresis. These data indicate that Pd nanoparticles were filled in nano pores of AAO substrate. This membrane has separation factor of hydrogen $\alpha = 3.85$.

Conclusions

In summary, we developed new preparation procedure which could make the very thin hydrogen perm-selective membrane. The procedure consisted of two preparation steps, one was the preparation of nanoparticles by sonochemical reduction and the other was deposition of the particles on a substrate by the electrophoresis method. The hydrogen permeance rate and hydrogen-nitrogen separation factor of the obtained membrane showed very high performance at the room temperature.

References

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2. K. Okitsu et al, *Chem. Mater.* 8, 315-317 (1996).