

Performance Evaluation of Hydrogen Permeability of pore-filling Membrane Prepared from Nanoparticles.

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Abstract

Pore-filling hydrogen perm-selective membranes were prepared with Pd nanoparticles that were made from Pd²⁺ ions by sonochemical reduction. The nanoparticles then deposited on a substrate disc with electrophoresis technique. The pore-filing membranes had high performance of perm-selectivity for H₂ with separation factor $\alpha=3.85$ under room temperature. It was suggested that the hydrogen molecules passed through the pore filling membrane via Knudsen diffusion process.

Introduction

To realize the H₂ permeable membrane with high permeance rate, thinner metallic membrane should be required. It's very difficult to make the H₂ permeable membrane thinner than 1 μm although many researchers try to make it using many techniques such as electric plating, non-electric plating, sputtering, etc. To prepare thin metallic membrane, we developed new procedure that consisted of sonochemical process and electrophoresis technique. The procedure was applied to make Pd metallic membrane with H₂ selective permeation. In this study, the effect of initial Pd²⁺ concentration for sonication on the H₂ permeability under room temperature will be reported. We also consider the permeation mechanism by the H₂ perm-selectivity.

Experimental

Pd nanoparticles were prepared from Pd²⁺ solution (0.1-1.0 mM) with ultrasonic reduction under Ar atmosphere²⁾. Formation of nanoparticles was confirmed by UV-vis spectra. Morphology of nanoparticles was observed by high-resolution transmission electron microscope (HR-TEM). From TEM photofraphs, average Pd particle size of each Pd concentration was calculated.

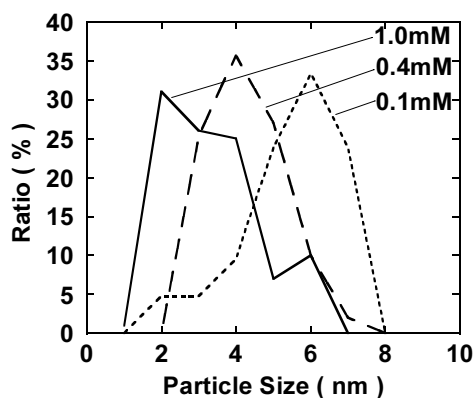


Fig. 1. Size distribution profiles of Pd nanoparticles prepared with sonochemical

The prepared Pd nanoparticles were deposited on anodic aluminum oxide substrates by electrophoresis. The morphologies and composition of the deposited membrane were observed by field emission scanning electron microscope (FE-SEM) and X-ray (EDS) analysis.

Permeations of N₂ and H₂ were evaluated by monitoring the pressure on both sides of membrane using a vacuum line under ambient temperature. Volume of permeation (q [mol]), permeability coefficient (R [mol / (m²·s·Pa)]), and separation factor (α) were calculated by equations described as follows,

$$q = P(p_1-p_2)A\Delta t / l, \quad R = P / l, \quad \alpha = R_{H_2} / R_{N_2},$$

where P is transmission coefficient [mol / (m·s·Pa)] (p_1-p_2) is differential pressure [Pa], A is membrane area [m²], Δt is differential time [s], l [m] is the membrane thickness.

Results and Discussions

Table 1 Gas permeation properties of prepared membranes

Initial Pd conc. [mM]	Average particle diameter [nm]	R [mol / (m ² ·s·Pa)]		α [-] (= R_{H_2}/R_{N_2})
		H ₂	N ₂	
0.1	5.1 ± 1.6	1.60×10 ⁻⁴	4.90×10 ⁻⁵	3.27
0.4	4.0 ± 1.3	8.00×10 ⁻⁶	2.08×10 ⁻⁶	3.85
1.0	3.0 ± 1.6	-	-	-

Pd nanoparticles prepared by different Pd²⁺ ion concentrations shows different UV-vis spectra and particle diameter. From Fig. 1, Pd particle diameter was proven to be different respectively depending on the concentration. The perm-selectivity of Pd membranes prepared from each concentration was compared in Table 1. The perm-selective performance were different according to the concentration, in other word, particle diameter affected the performance. We will discuss the mechanism of gas permeation by use of membrane thickness and simulation, in this presentation.

Conclusions

In summary, the hydrogen permeation rate and hydrogen-nitrogen separation factor of the obtained membrane showed very high performance at room temperature. As a result of consideration, the H₂ permeation mechanism depended on the difference between Pd concentration and the particle diameter. The preparation method of this hydrogen perm-selective pore-filling membrane has potential to spread widely

References

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