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Polarization phenomena of hydrogen-rich gas in high-permeance Pd and Pd–Cu membrane tubes

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HIGHLIGHTS

- Concentration polarization phenomena in membrane tubes are studied.
- Two pure palladium (Pd) membranes and one Pd–Cu alloy membrane are tested.
- Concentration polarization is clearly exhibited when the gas mixture is fed.
- Decreasing H_2 concentration in the feed gas deteriorates polarization.
- For tested gas mixtures, the permeance is reduced by 1–2 orders of magnitude.

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ABSTRACT

Concentration polarization, a retarding effect upon hydrogen permeation across a membrane, will affect the performance of hydrogen separation. To recognize the concentration polarization behavior of hydrogen-rich gas in high-permeance membrane tubes, three palladium-based membranes with two pure palladium (Pd) membranes and one Pd–Cu alloy membrane are tested. Three important parameters, consisting of the H_2 partial pressure difference, the flow rate at the exit of the retentate side and the H_2 concentration in the feed gas mixture (H_2+N_2), are taken into account. A comparison to pure H_2 permeation suggests that the concentration polarization is clearly exhibited when the gas mixture is fed. Decreasing the H_2 concentration in the feed gas tends to deteriorate the polarization phenomenon. The sensitivity of polarization to H_2 concentration is higher when its concentration in the feed gas is larger. For the gas mixtures with 50 and 75 vol% of H_2, the permeances of the membranes are reduced by 1–2 orders of magnitude. The permeances of the membranes are also affected a bit when the H_2 partial pressure difference or the flow rate is altered.

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1. Introduction

Fuel cells are a potential device for prospective power generation because of their high efficiency of energy conversion. Among developing fuel cells, proton exchange membrane fuel cells (PEMFCs), which belong to low-temperature fuel cells, have received much attention compared to others. This arises from the fact that PEMFCs can be extensively applied for producing power in vehicles, residences and portable devices [1]. The required feedstock for driving PEMFCs is high-purity hydrogen. Consequently, the mass production of high-purity hydrogen is of the utmost importance when PEMFCs are employed widely.

Thermochemical methods, such as steam reforming, partial oxidation, autothermal reforming and water gas shift reaction, are the crucial and commonly utilized routes for the generation of hydrogen [2–4]. For example, the steam reforming of natural gas has been widely practiced in industry to produce hydrogen [5]. However, the hydrogen-rich gas rather than pure hydrogen is obtained from the fuel processing. It is essential to separate and purify hydrogen from the hydrogen-rich gas so as to drive PEMFCs. Hydrogen separation and purification via palladium (Pd) and Pd-alloy membranes have been regarded as an important way to fulfill hydrogen production lately [6–9]. This is attributed to the fact that hydrogen separated by Pd-based membranes possesses some advantages over other methods, say, pressure swing adsorption and cryogenic distillation. Pd-based membranes are characterized by long lifetime, low investment cost, low energy consumption and high hydrogen selectivity [10]. They are also more durable to withstand thermal cycling. High purity hydrogen obtained from the applications of Pd-based membranes can be used directly in PEMFCs. For these reasons, Pd-based membranes have been thought of as a proper candidate for producing hydrogen

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Nomenclature

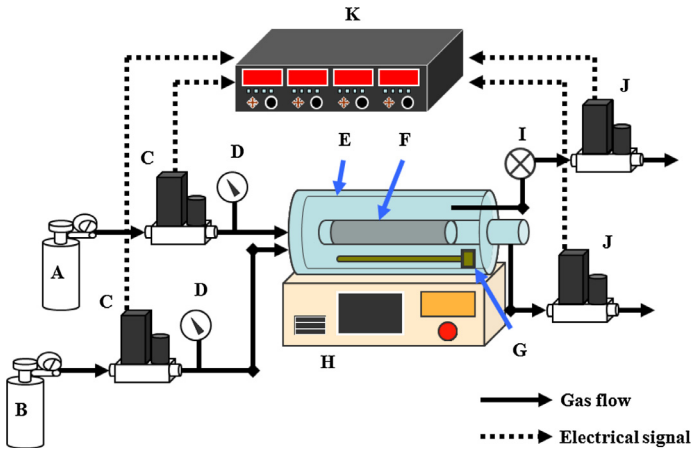
A	membrane surface area (m^2)
K	permeance (mol m^{-2} s^{-1} Pa^{-n})
K'	permeability (mol m^{-1} s^{-1} Pa^{-n})
l	thickness of membrane (m)
$\dot{m}$	permeation rate (mol s^{-1})
n	pressure exponent (–)
P	pressure (Pa)
Q	flow rate (cc min^{-1})
T	temperature (K)
Subscript	
H_2	hydrogen
r	retentate side
p	permeate side

with high purity, especially in small-scale and moderate-scale facilities.

Hydrogen permeation through a Pd-based membrane is mainly governed by a chemisorption–dissociation–diffusion mechanism [11,12]. Specifically, the processes include: (1) dissociatively adsorbing H_2 onto metal surface; (2) diffusing atomic H through bulk metal; and (3) associatively desorbing H_2 from the metal surface [13]. In general, hydrogen permeation across a dense Pd-based membrane is controlled by the diffusion of atomic hydrogen in the membrane. That is, the mass transfer of hydrogen atoms is the rate-limiting step in the entire H_2 separation process. This is the reason that Sieverts' law has been extensively employed to describe H_2 flux across a membrane.

Generally, the denser the Pd-based membrane, the higher the purity of H_2 at the permeate side of the membrane. However, this may result in a lower H_2 flux. In other words, it seems to be inevitable in reducing permeation rate from the penalty of gaining high-purity H_2. As a matter of fact, when H_2 is used in PEMFCs, the prime concerned gas in the feedstock is CO in that it will poison the platinum electrodes in the fuel cells [14,15]. Instead of dense membranes, if high-permeance Pd-based membranes are developed to treat hydrogen-rich gas, more H_2 can be obtained. Besides, high-permeance Pd-based membranes are usually accompanied by thinner membranes, implying that the cost of manufacturing membrane tubes can be lower. Though little amount of CO may also permeate through the membranes, it can be eliminated by means of subsequent methanation [16] or preferential oxidation (PROX) [17]. Accordingly, using high-permeance Pd-based membranes in association with CO removal is a feasible alternative to gain the feedstock of PEMFCs.

In reviewing past studies, it can be found that concentration polarization is a noticeable factor in retarding hydrogen permeation [18,19]. Concentration polarization comes from the elicited concentration boundary layer along the membrane surface which results in



A: nitrogen cylinder; B: feed gas cylinder; C: electronic flow rate meter;
D: pressure gauge; E: electric furnace; F: membrane tube; G: thermocouple;
H: power controller; I: back pressure valve; J: electronic flow rate meter; K: readout

Fig. 1. A schematic of experimental system.

Table 1
A list of membrane characteristics.

Membrane	Membrane/support	Thickness (μm)	Membrane area (m^2)	Hydrogen flux[a] (mol m^{-2} s^{-1})	Permselectivity (H_2/N_2)
A	Pd/PSS	6.93	0.00424	0.785483	243.84
B	Pd–Cu/PSS	7.03	0.00424	0.754796	888.091
C	Pd/PSS	6.63	0.00424	0.596061	1678.73

[a] Hydrogen flux is measured by pure hydrogen under the pressure difference of 4 atm.

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