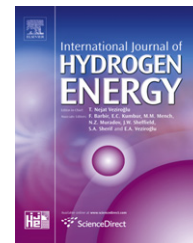


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Numerical characterization on concentration polarization of hydrogen permeation in a Pd-based membrane tube

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ABSTRACT

Sieverts' law has been extensively employed to evaluate hydrogen permeation rate across a hydrogen-permeable membrane based on the concept of continuous stirred tank reactor (CSTR). However, when the hydrogen permeation rate is high to a certain extent, concentration polarization will appear in a membrane tube which results in the deviation of hydrogen permeation rate from Sieverts' law. Under such a situation, the nature of mass transfer in a membrane tube is characterized by plug flow reactor (PFR) rather than CSTR. To figure out the feasibility of Sieverts' law, a two-dimensional numerical method is developed to simulate the phenomena of concentration polarization for hydrogen permeation in a Pd-based membrane tube. Four important parameters affecting hydrogen permeation are taken into account; they include the pressure difference, H₂ molar fraction in the influence, Reynolds number and membrane permeance. The predictions indicate that increasing pressure difference or membrane permeance facilitates H₂ permeation rate; concentration polarization is thus triggered. Alternatively, when Reynolds number or H₂ molar fraction decreases along with a higher permeance, the deviation of PFR from CSTR grows, even though H₂ permeation rate declines. From the obtained results, it is concluded that the H₂ permeation rate can be predicted by Sieverts' law if the H₂ permeation ratio is no larger than 30%.

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1. Introduction

Hydrogen production is an important process in refineries and petrochemical industry; it also plays a crucial role for the prospective hydrogen economy because of possibly extensive applications of low-temperature fuel cells, say, proton exchange membrane fuel cells (PEMFCs). PEMFCs, using pure hydrogen as the fuel, are likely to be used in transportation, stationary power systems and portable electronic devices. Pure hydrogen can be obtained from electrolysis, water splitting and thermal decomposition of methane [1–3].

Nevertheless, these methods still suffer from more energy consumption, low efficiency or relatively high cost. In contrast, mass production of hydrogen-rich gases has been widely practiced through thermochemical routes, such as steam reforming (SR) and gasification [4–6]. In addition to the aforementioned two methods, the production of hydrogen-rich gases via autothermal reforming (ATR), partial oxidation (POX) and pyrolysis have also received a great deal of attention [7,8]. The produced hydrogen-rich gases can be further processed through water gas shift reaction (WGS) to enrich hydrogen [9]. Once the hydrogen-rich gases are generated,

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Nomenclature		w	Velocity (m s^{-1})
A_m	Surface area of membrane area (m^2)	x_i	Molar fraction of species i (dimensionless)
A_{pre}	Pre-coefficient ($\text{mol m}^{-1} \text{s}^{-1} \text{Pa}^{-n}$)	<i>Greek letters</i>	
B	Diffusion factor ($\text{Pa m}^2 \text{s}^{-1}$)	δ	Membrane thickness (m)
c	Molar concentration (mol m^{-3})	μ	Viscosity (Pa s)
c_p	Gas mixture specific heat ($\text{J kg}^{-1} \text{K}^{-1}$)	ρ	Gas mixture density (kg m^{-3})
D	Diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	Φ	Binding factor (dimensionless)
E_a	Activation energy (J mol^{-1})	<i>Subscript</i>	
F	Flux ($\text{mol m}^{-2} \text{s}^{-1}$)	H_2	Hydrogen
K	Permeance ($\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-n}$)	i	Species i
k	Fluid phase thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	in	Inlet
M_i	Molar mass of species i (kg mol^{-1})	j	Species j
p	Pressure (Pa)	N_2	Nitrogen
R	Universal gas constant ($8.314 \text{ m}^3 \text{Pa K}^{-1} \text{mol}^{-1}$)	p	Permeate side
T	Temperature (K)	r	Retentate side
V	Velocity (m s^{-1})		

how to separate and purify hydrogen from the mixtures for low-temperature fuel cells, refineries or petrochemical processes becomes an essential and vital task. In the currently available techniques, pressure swing adsorption, cryogenic distillation and solvent adsorption [10] have been utilized for hydrogen separation. But these procedures are relatively expensive. Another feasible method is membrane separation which provides a number of advantages over the other separation technologies, such as lower cost, fewer moving parts as well as high H_2 selectivity and purity.

Among the materials of membrane, palladium (Pd) has been considered as the most important material for hydrogen separation in that Pd membranes possess high permeability and infinite selectivity of H_2 as well as chemical compatibility [11–14]. For this reason, Pd has been widely applied in membrane separation technology. Unfortunately, pure Pd membranes cannot be used for hydrogen separation below $300 \text{ }^\circ\text{C}$ [12]. This arises from hydrogen embrittlement of Pd layer due to the lattice expansion caused by dissolved hydrogen [15,16]. In order to reduce the cost and improve the properties of pure Pd membranes, some Pd-alloy membranes, say, Pd–Au, Pd–Ag, Pd–Ni and Pd–Cu [17,18], coating upon a variety of supports have been developed in the last decade. For example, it was reported that Pd–Ag alloys were found which show the embrittlement resistance and greater permeability [18].

When hydrogen is separated from a hydrogen-rich gas by a membrane, its permeance plays a key role in determining the performance of the membrane. Once the permeance of a membrane is measured, the hydrogen flux across the membrane can be predicted by

$$F_{\text{H}_2} = K \left(P_{r,\text{H}_2}^n - P_{p,\text{H}_2}^n \right) \quad (1)$$

In the above equation, K is the permeance of the membrane; P_{r,H_2}^n and P_{p,H_2}^n stand for the hydrogen partial pressures at the retentate side and the permeate side of the membrane, respectively. In general, the value of the pressure exponent n is between 0.5 and 1 [19]. In the entire chemisorption-dissociation-diffusion process, if the surface reaction process is the rate-controlling step, the hydrogen flux can be

evaluated using $n = 1$ [20–22]. Alternatively, once the diffusion of atomic hydrogen through the dense metal layer is the rate-limiting step, it is proper to use $n = 0.5$ and Eq. (1) becomes well-known Sieverts' law. That is

$$F_{\text{H}_2} = K \left(P_{r,\text{H}_2}^{0.5} - P_{p,\text{H}_2}^{0.5} \right) \quad (2)$$

Sieverts' law has been extensively employed to evaluate hydrogen permeation rate across a hydrogen-permeable membrane using the concept of continuous stirred tank reactor (CSTR). That is to say, it is assumed that the hydrogen partial pressures at the retentate side and the permeate side are uniform. However, when a hydrogen-rich gas flows through a membrane tube, the mass transfer process pertains to a plug flow reactor (PFR) in nature, especially at the situation of high hydrogen permeation rate. As a result, the phenomenon of concentration polarization will occur along the membrane surface [23,24]. Specifically, when gas mixture is brought to a membrane surface, a depletion of permeable component in the boundary layer near the membrane surface causes a concentration gradient which has a serious adverse effect on the performance of a membrane-based separation unit. This phenomenon is referred to as the concentration polarization [24]. Under such a situation, the hydrogen flux departs from the prediction of Sieverts' law, that is, the law fails in evaluating hydrogen permeation rate.

To recognize the behavior of concentration polarization in a membrane, a number of studies have been carried out. For example, Ludtke et al. [25] experimentally and theoretically investigated the effect of concentration polarization on the separation of gas-vapor mixtures. He et al. [26] developed a mathematical model to analyze the concentration polarization of gas separation. Mourgues and Sanchez [23] also developed a mathematical model to explore the concentration polarization in gas separation hollow-fibers modules. Zhang et al. [24] paid their attention to the concentration polarization in H_2 enrichment by highly permeable and selective Pd membranes using both the experimental and numerical approaches. In the experimental study of Pizzi et al. [27], H_2/N_2 and H_2/CO mixtures separated by Pd–Ag membranes under the effect of concentration polarization were tested. From

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