Journal of Membrane Science 472 (2014) 45–54

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Flow-field design for improving hydrogen recovery in a palladium membrane tube

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ARTICLE INFO

Article history:
Received 17 April 2014
Received in revised form
16 August 2014
Accepted 24 August 2014
Available online 2 September 2014

Keywords:
Palladium membrane
Concentration polarization
Hydrogen recovery
Flow-field design
Numerical simulation

ABSTRACT

The installation of baffles in a membrane tube is an effective way to lessen concentration polarization and improve hydrogen separation efficiency. In order to enhance the performance of hydrogen recovery, the flow-field design in a membrane system, which is composed of an internal tube and a shell, is performed through the computational fluid dynamics. The influences of baffle pattern, baffle location, and the ratio of baffle length to shell radius ($L/0.5D_0$) on hydrogen recovery are analyzed. The impacts of the Reynolds numbers in the tube and the shell as well as the pressure difference across the tube on hydrogen permeation are also evaluated. The predictions suggest that a single baffle installed on the shell and located at the leading edge of the membrane can effectively improve hydrogen separation. Increasing the ratio of $L/0.5D_0$ and decreasing the Reynolds number of feed gas intensify hydrogen recovery in a significant way, whereas increasing the Reynolds number of sweep gas merely raises the recovery to a small extent. Installing a baffle in the membrane tube enables the increment of hydrogen recovery up to 10% when compared to that without baffle. Hydrogen in the feed gas can be recovered completely at $L/0.5D_0=0.75$ when the Reynolds number of the feed gas is as low as 15.

1. Introduction

Hydrogen, the most abundant element in the universe [1], is considered as an important energy carrier or fuel for prospective transport and power systems. However, most of the hydrogen combines with carbon and oxygen and is stored in biomass, hydrocarbons, or water [2,3], fuel processing for extracting hydrogen from these compounds is thus essential. Currently, a number of thermochemical methods, such as, steam reforming [4], partial oxidation [5], autothermal reforming [6,7], water gas shift reaction [8], and gasification [9], can be adopted to produce hydrogen. In addition to hydrogen, the gas products from these methods also contain carbon monoxide, carbon dioxide, and other low-concentration gases. The processes of hydrogen separation, say, pressure swing adsorption, cryogenic distillation, and membrane separation, are thus required to purify hydrogen from hydrogen-rich gases [10,11].

As far as hydrogen separation by membranes is concerned, the studies of palladium-based (Pd-based) membranes command both fundamental and practical interest. On the fundamental side, because of the catalytic effect of palladium on hydrogen, hydrogen permeation through a Pd-based membrane essentially involves a number of chemical and transport mechanisms. Specifically, the permeation process proceeds from hydrogen dissociation, the diffusion of atomic hydrogen, and then to the association of atomic hydrogen [12]. On the practical aspect, Pd-based membranes possess the advantages of high selectivity and permeability of hydrogen, easy operation and maintain, long lifetime, low energy consumption, and lower facility cost [13–15]. Therefore, hydrogen separated by Pd-based membranes is a promising route to produce pure hydrogen for fuel cell applications, especially for small-scale production and on-board utilization [16–18]. Moreover, after undergoing hydrogen separation, the CO_2 concentration in the treated gas is enriched. This is conducive to subsequent CO_2 capture, thereby abating greenhouse gas emissions into the atmosphere [19,20].

When hydrogen permeates through a Pd-based membrane, its flux (mol m^{-2} s^{-1}) across the membrane is commonly described by

$$F_{H_2} = K\left(p_{r,H_2}^{n} - p_{p,H_2}^{n}\right) \quad (1)$$

where K and n are the permeance (mol m^{-2} s^{-1} Pa^{-n}) and pressure exponent, respectively, and the value of n is usually between 0.5 and 1 [21]. In practice, the higher the hydrogen recovery, the better the performance of the membrane is. However, a high hydrogen permeation rate may be accompanied by the phenomenon of concentration polarization, stemming from the development of hydrogen concentration boundary layer and the attenuation of hydrogen concentration along the membrane surface at the retentate side [22]. If the hydrogen

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http://dx.doi.org/10.1016/j.memsci.2014.08.041
0376-7388/

concentration boundary layer is disturbed or even destroyed, the concentration polarization will be lessened, thereby enhancing hydrogen permeation rate.

The disturbance or destruction of hydrogen concentration boundary layer along the membrane surface can be implemented through the flow-field design. The installation of baffles in membrane tubes may be an effective means to achieve the flow-field design. In reviewing past studies concerning membrane systems with baffles, very few studies [23–26] have been performed. Liu et al. [23] studied the removal of volatile organic compounds from water using a pervaporation process with baffle installation. They found that installing baffles were conducive to the separation process and an increase in the height of baffle improved the mass flux. He et al. [24] designed two different installation patterns of baffles, namely, the central baffles and the wall baffles, to investigate their influences on the performance of filtration, and reported that either central or walls baffles caused intensive fluctuations of local wall velocity and shear stress, thereby enhancing the filtration performance. Coroneo et al. [25] studied hydrogen separation from a CH_4–CO_2–H_2–H_2O gas mixture in a Pd–Ag membrane system at various numbers of baffles, and discovered that increasing baffle number advanced the hydrogen separation efficiency. Chen et al. [26] numerically investigated hydrogen separation from a H_2–CO–CO_2 gas mixture at baffle numbers of 0–4 where steam was used a sweep gas to facilitate hydrogen permeation at the permeate side. They addressed that the higher the number of baffles, the better the hydrogen recovery.

From the literature review described above, only two studies [25,26] concerning hydrogen separation by Pd-based membranes with baffles were performed, and there still remain a lot of hydrogen transport mechanisms that are not recognized in sufficient detail. Over the past several decades, on account of much progress in computer capabilities and numerical algorithms, numerical simulation has become a powerful tool to predict physical phenomena and can aid in designing reactors [27–29]. In a previous study [30], a numerical method considering conjugate hydrogen permeation and polarization in a Pd membrane tube was developed. The influences of the flow patterns of feed gas and sweep gas as well as their flow rates on hydrogen separation were investigated. Alternatively, the influence of baffle installation on hydrogen separation was preliminarily studied [26]; however, only the effect of the number of baffles on H_2 recovery was considered. To provide a comprehensive study on the improvement of hydrogen recovery under the impact of baffle installation, a number of factors affecting H_2 recovery, such as, the baffle pattern, baffle location, and the ratio of baffle length to shell radius, are taken into account in the present study. Particular attention is paid to the flow-field design to minimize concentration polarization and intensify hydrogen permeation rate and recovery ratio.

2. Mathematical formulation

2.1. Permeation system and assumptions

Fig. 1a shows the geometry of the membrane system. The system is a concentric tube configuration which consists of an internal tube (100 mm i.d.) and a shell (8 mm i.d.). The lengths of the tube and the shell are 400 mm, while the length of the membrane is 150 mm. The feed gas and the sweep gas are sent into the shell and the internal tube, respectively [26]. The physical phenomena in the present study are subject to the following assumptions:

(1) The hydrogen permeation process is steady and isothermal (at 350 °C);
(2) the flow fields in the tube and the shell are laminar and axisymmetric;
(3) the permselectivity of the membrane to hydrogen is infinite, whereas its permselectivities to other gases are zero; and
(4) the gas mixture is well-mixed at the inlet, and the gases at the permeate side and the retentate side abide by the ideal gas law.

2.2. Modeling, equations, and boundary conditions

To deal with the physical phenomena of hydrogen permeation across a Pd-based membrane, the concept of conjugate hydrogen permeation [30] is adopted to approach hydrogen permeation process; that is, the fluid dynamics and mass transfer at the both sides of the membrane are fully simulated. To approach the hydrogen permeation in the membrane, it is conceived to be composed of two zones, with a hydrogen-sink zone and a hydrogen-source zone [25] at the retentate side and the permeate side, respectively. Accordingly, the entire computation domain is partitioned into four domains: (1) the tube (Domain A); (2) the internal membrane or hydrogen-source region (Domain B); (3) the external membrane or hydrogen-sink region (Domain C); and (4) the shell (Domain D), as shown in Fig. 1b.

On account of the four domains encountered, four sets of the conservative equations, including continuity, momentum, and species equations as well as the equation of state are invoked to describe mass, momentum, and species transports in the system. The aforementioned equations are shown in Table 1. In the tube and the shell (i.e. Domains A and D), no source or sink terms are considered. In the internal and external membranes (i.e. Domains B and C), the source or sink terms of gases, except for hydrogen, are disregarded; with regard to hydrogen, a source-sink pair is utilized and hydrogen permeation in the source and sink terms obey Sieverts' law. Details of the source and sink terms are tabulated in Table 2. In the entire computational domain, the boundary conditions are made up of: (1) the upstream inflow; (2) the downstream outflow; (3) the tube centerline; (4) the tube wall and the membrane centerline; (5) the shell wall; (6) the interface between the shell (Domain D) and the sink domain (Domain C) and (7) the interface between the internal tube (i.e., Domain A) and the source domain (Domain B). Details of the boundary conditions are sketched in Fig. 1b.

2.3. Properties of gas mixture

In the permeation system, four different gases, H_2, CO, CO_2, and H_2O, are simultaneously considered to account for hydrogen separation from a gas mixture. Wilke semi-empirical correlation [31–33] is adopted to evaluate the viscosity of gas mixture and the correlation is expressed as

$$\mu = \sum_{i=1}^{N} \frac{x_i \mu_i}{\sum_{j=1}^{n} x_j \phi_{ij}} \tag{2}$$

When calculating the binary mixture diffusivity of gas mixture under an isothermal condition, the Chapman–Enskog equation [32,33] is considered and it is written as

$$D_{ij} = 1.881 \times 10^{-3} \times \frac{T^{1.5}\left((1/M_i)+(1/M_j)\right)^{0.5}}{p\sigma_{ij}^2 \Omega_d} \tag{3}$$

where T, p, σ_{ij} and Ω_d are temperature, absolute pressure, interaction value for binary mixture and diffusion collision integral, respectively. The diffusion collision integral Ω_d is correlated by

$$\Omega_d = \frac{A}{\varsigma^B} + \frac{C}{\exp(D\varsigma)} + \frac{E}{\exp(F\varsigma)} + \frac{G}{\exp(H\varsigma)} \tag{4}$$

$$\varsigma = \frac{\overline{k}T}{\varepsilon_{ij}} \tag{5}$$