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Preparation and characterization of inorganic membranes for hydrogen separation

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ARTICLE INFO

Article history:
Received 16 July 2015
Received in revised form
12 November 2015
Accepted 17 November 2015
Available online 6 February 2016

Keywords:
Tubular alumina support
Silica membrane
Pt membrane
Hydrogen selectivity

ABSTRACT

A tubular commercially available alumina support was coated using the dip coating technique. The objective is to prepare silica and Pt impregnated membranes. Scanning electron microscopy (SEM), energy diffraction X-ray analysis (EDXA), nitrogen adsorption–desorption at 77 K and gas permeation measurements were employed for membrane characterization. The permeation of H_2, He and N_2 revealed that the membranes are crack-free. H_2/N_2 selectivity for the silica membrane obtained the highest value of 2.93 at 0.9 barg and 25 °C. On the other hand, H_2/He selectivity of 1.96 at 1.6 barg and 300 °C for the Pt membrane was obtained and found to be higher than the theoretical Knudsen selectivity. While the silica membrane realised on the thin film coating to enhance the selectivity to hydrogen, the Pt impregnated membrane on the other hand enhance hydrogen transport through an activated surface diffusion in addition to Knudsen flow.

Introduction

Hydrogen is currently mainly used in the chemical industry for the chemical production of methanol and ammonia, and for cleaner transportation fuels [1]. Hydrogen is anticipated to replace fossil fuels in the near future, and thus significantly contribute to the atmospheric air quality [1]. The increasing demand for cleaner energy has resulted in the global need to adopt the projected hydrogen economy as the key possible long-term solution to the growing energy crises [2]. In recent years, the use of conventional fossil fuel sources has increased as a transitional measure towards hydrogen economy where coal gasification is considered as dominating the process in delivering hydrogen due to its enormous reserves which is speculated to last for at least 50 decades [3].

Literature shows that hydrogen can actually be separated with inorganic membranes [4–8]. Inorganic membranes derived from silica, ceramics and metal alloys are candidates for high temperature gas separation. Palladium (Pd) [4,9], platinum (Pt) [9] and their alloys are the ideal membranes applied for high purity hydrogen production from mixed gas streams even though these metals are expensive [4,9,10]. Pd-based membranes are attractive for membrane reactor applications because dense Pd is highly permeable to hydrogen and if properly configured can offer better thermal stability and selectivity than polymer and microporous membranes [9]. Transport of hydrogen through dense Pd membranes follows the solution diffusion mechanism where only hydrogen is transported resulting in high purity (99.9999%), but have been limited in commercialization due to issues which include support quality, surface poisoning due to carbon species,

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http://dx.doi.org/10.1016/j.ijhydene.2015.11.179

Table 1 – Characteristics of ceramic membranes.

Membrane	Operating temperature (K)	Pore diameter (nm)	Internal diameter (mm)	Outer diameter (mm)	Porosity (%)	Length (mm)	Area (m^2)	Wall thickness (μm)
Alumina support	298–573	4.17	7	10	45	348	0.0062	–
Silica membrane	298–573	3.94	7	10	–	348	0.0062	–
Platinum membrane	298–573	3.70	7	10	45	348	0.0062	10.97–12.55

hydride formation, and irreversible damage caused by bulk sulfide formation [9,11,12].

Sol–gel method has been proposed by many researchers as the ideal technique for membrane preparation [8]. This technique has many merits for preparing pore separation layer on the support [8]. Silica membranes are among the candidates for low-cost hydrogen separation and purification [2,3]. In fact, these membranes can accommodate the separations of hydrogen, nitrogen, carbon dioxide, helium and oxygen. The main characteristics of inorganic membranes are permeance and selectivity or separation factor [13]. Permeance is a measure of the gas flow rate per unit area per unit pressure difference. Permeance is a more practical unit than permeability because the thickness of the membrane in most cases is not known very accurately [13]. Permeance of gas is therefore defined as;

$$F = q/A\Delta P \quad (1)$$

where F is the Permeance (mol/m^{-2} s^{-1} Pa^{-1}), q is the molar flow (mol/sec), A is the surface area of the membrane (m^2), ΔP is the pressure difference across the membrane (Pa).

Permeability of gas is defined as the permeance multiplied by the thickness of the membrane and is written as;

$$P_e = L \times F \quad (2)$$

where P_e is the Permeability (mol-m/m^{-2} s^{-1} Pa^{-1}) and L the thickness of the membrane (m).

The selectivity is defined as the ratio of the pure component permeabilities (P_y and P_z) for single gases. It can be written as;

$$\alpha_{y,z} = P_y/P_z \quad (3)$$

where P_y is the permeability of y component (mol-m/m^{-2} s^{-1} Pa^{-1}), and P_z is the permeability of z component (mol-m/m^{-2} s^{-1} Pa^{-1}).

The transport of gases through membranes behaves differently as the pore diameter is reduced. Gas transport can be affected by pressure and temperature. A change in

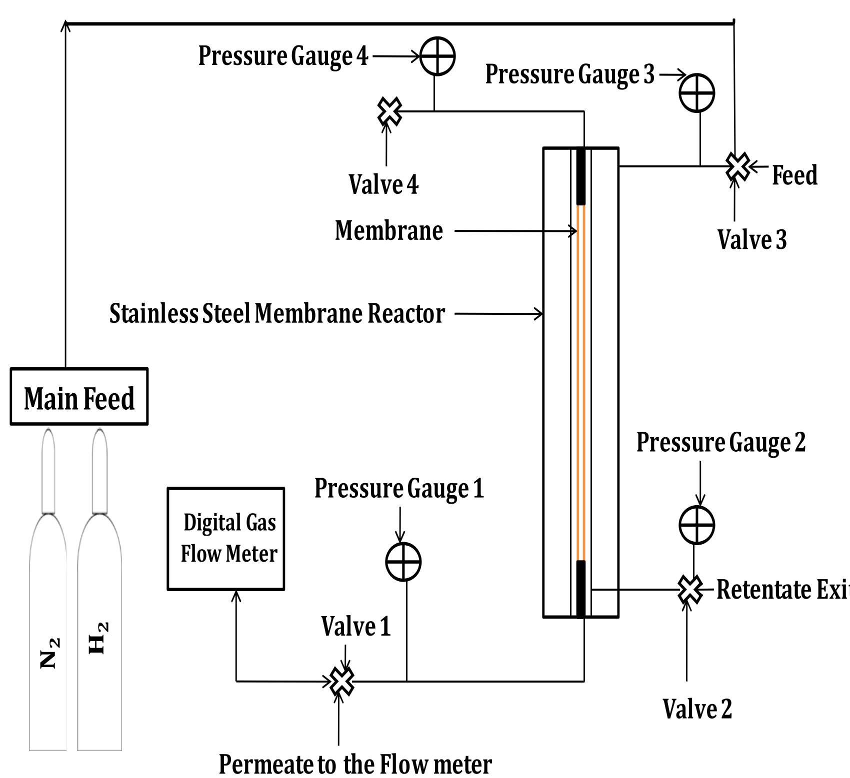

Fig. 1 – Schematic diagram of the experimental setup.