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Hydrogen Separation Using Inorganic Membranes

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Abstract

Gas permeation of hydrogen (H_2) and nitrogen (N_2) were obtained from 30 and 6000 nm pore diameter tubular commercial alumina ceramic membranes at 0.05 to 1.00 bar and 298 K. Flow rates of up to 3.279 and 2.296 l/min were obtained for H_2 and N_2 respectively. The ratio of H_2/N_2 flow rates were used to calculate H_2/N_2 selectivity. The experimental H_2/N_2 selectivities obtained were 1.85 and 1.43 for the 30 and 6000 nm respectively.

Introduction

Almost 80 percent of the global energy demand comes from fossil fuels such as natural gas, coal among others. Unlike using fossil fuels, hydrogen when combusted produces only water as byproduct [1]. The world energy consumption is forecasted to rise by 56 percent from 2010 to 2040 mainly from coal and natural gas [2]. Therefore, it is advantageous to substitute hydrogen with the current fossil fuels because it is widely accepted as a clean energy carrier in for instance fuel cell systems. These could help to address the problems linked to energy security which includes air pollution and global climate change. The need for hydrogen as a source of renewable energy will be enhanced in the coming years due to its demand for raw material processing in the chemical industry as well as home heating [3]. The state of the art membrane technology has emerged as an attractive substitute in the chemical industry, particularly for hydrogen separation from process streams.

Membranes can be classified into inorganic and organic/inorganic (hybrid) systems. The organic ones are further divided into biological and polymeric constituents, while the inorganic membranes can be divided into metallic and ceramic (porous and non-porous) membranes [4]. The applications of inorganic membranes have received considerable attention during the past three decades in a wide array of industrial operations for gas separation due to their thermal, chemical and mechanical stability when compared with polymeric membranes. This includes hydrogen separation/recovery for fuel cell application to generate electricity [5], carbon dioxide separation from fuel gas [6], hydrogen sulphide separation from natural gas [7] and recovery of methane from bio-gas [5]. Inorganic membranes are environmentally benign, cost-effective and energy efficient.

According to the International Union of Pure and Applied Chemistry (IUPAC), porous membranes are classified as; Micropores 0.5 - 2 nm where separation is based on molecular sieving mechanism, mesopores 2 - 50 nm where Knudsen flow mechanism is the dominant flow but multilayer flow and/or capillary condensation and viscous flow can also take place, and macropores > 50 nm where there is no separation and the flow mechanism is basically influenced by viscous flow [8-10]. In all cases, some considerations on productivity and separation selectivity, mechanical integrity, and membrane's durability at the operating conditions needs to be taken into account against cost issues [4], because the importance of each of these requirements differs with their applications. Permeability and selectivity are the main basic properties of a membrane.

The main aim of the present paper is to report experiments undertaken using a tubular commercial alumina membrane to understand the transport through inorganic membranes for hydrogen separation from natural gas.

Experimental

Commercially available γ-alumina mesoporous ceramic membranes (77% alumina and 23% TiO_2) tubes 368 mm long with 7 and 10 mm internal and outer diameter where obtained from Ceramiques Techniques et Industrielles (CTI SA) France. The ceramic membranes had a 30 and 6000 nm pore diameter.

Figure 1 shows the permeation system. Each membrane was sealed in a stainless-steel reactor by graphite O-rings which are high temperature resistant. The feed pressure used was between 0.05 to 1.0 bar and was measured by a pressure transducer. At the permeate, the permeation flux was measured using a flowmeter, and a thermometer to record the required temperature. The permeate side was fully opened to the atmosphere. The temperature was maintained at 298 K. The permeate pressure was always the atmospheric one.

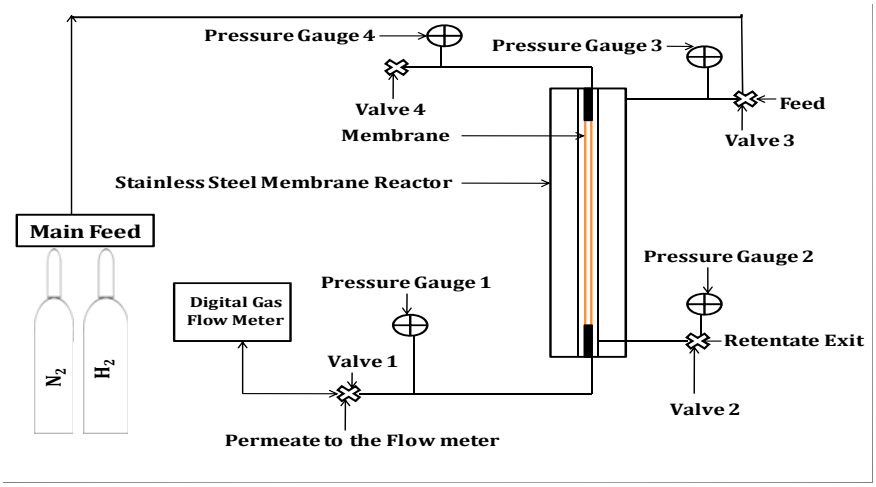

Figure 1: Permeation test experimental system.

Results and discussion

Figure 2 shows the H_2 and N_2 single gas flow rates as a function of feed pressure through a 30 and 6000 nm commercial alumina membrane was studied at 298 K. H_2 flow rates on 6000 nm permeated faster than that of 30 nm, and a similar trend can be seen for N_2 flow rates (Figure 3). This result indicates that the higher the pore size, the higher the permeation rate.

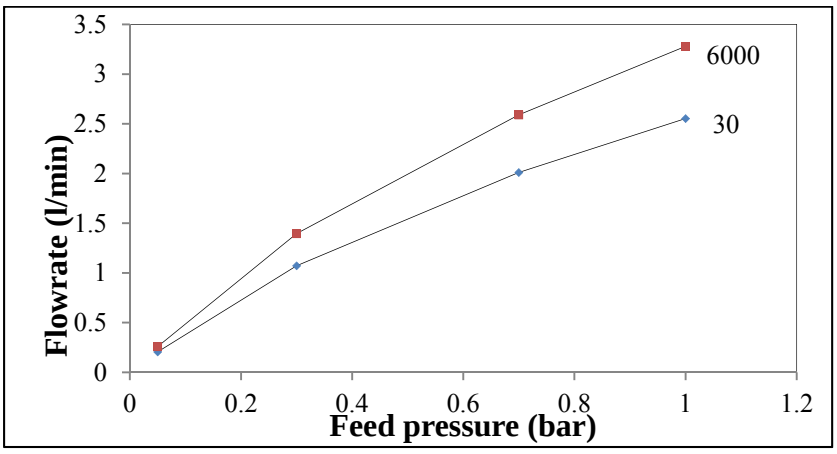

Figure 2. H_2 flow rate as a function of feed pressure for 30 and 6000 nm at 298 K.

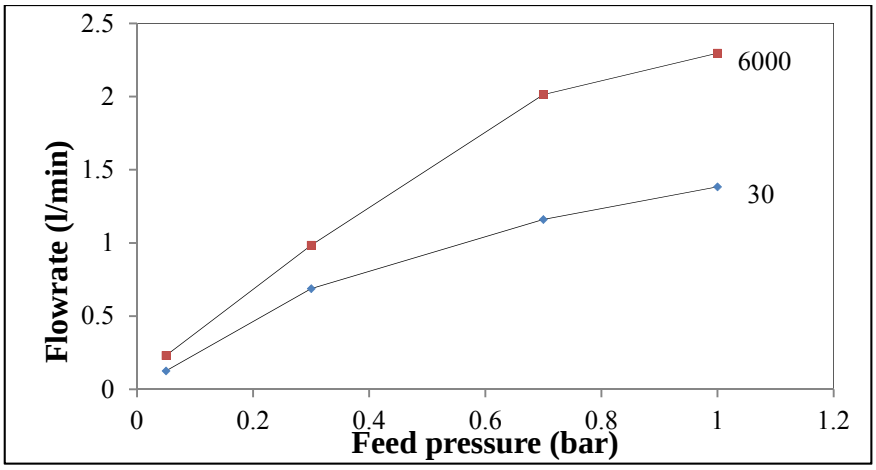

Figure 3. N_2 flow rate as a function of feed pressure for 30 and 6000 nm at 298 K.

Figure 4 shows a comparison of the H_2 and N_2 flow rates as a function of feed pressure for the 30 nm pore membrane. It can be seen that hydrogen had the higher flow rate due to its lower molecular weight compared with nitrogen which indicates knudsen flow contribution. Table 1 shows H_2 and N_2 flow rates and H_2/N_2 selectivities from 30 and 6000 nm commercial alumina membrane at 1 bar and 298 K. Experimental gas selectivities were calculated as the ratio of H_2 and N_2 flow rates. The experimental H_2/N_2 selectivities obtained were 1.85 and 1.43 from 30 and 6000 nm respectively. An increase in selectivity was obtained from 30 nm pore diameter membrane. This is a clear indication that the smaller the pore diameter, the higher the selectivity that would be achieved. Consequently, microporous membranes can be employed in order to enhance higher selectivities than those of knudsen.

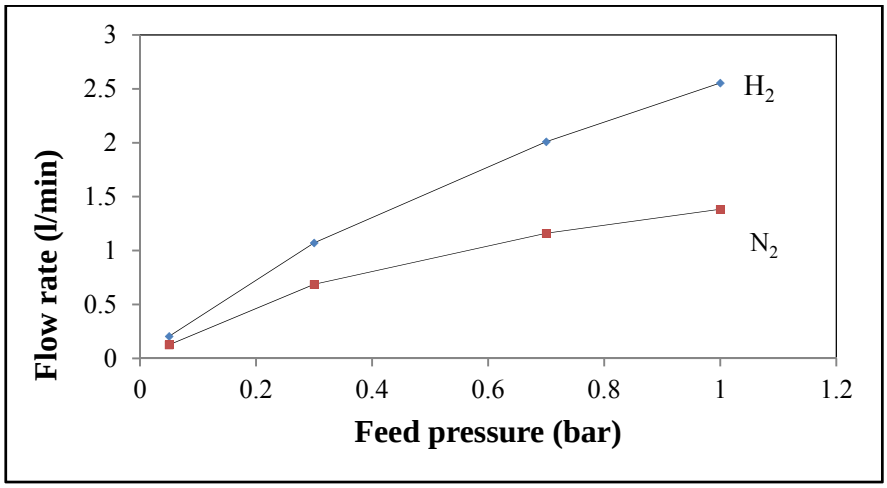

Figure 4. H_2 and N_2 flow rate as a function of feed pressure for 30 nm at 298 K.

Table 1. Hydrogen and nitrogen flow rates and α H_2/N_2 at 1 bar and 298 K

Membrane pore diameter (nm)	Flow rate (l/min)		Selectivity, α (H_2/N_2)	
	H_2	N_2	Theoretical	Experimental
30	2.554	1.384	3.74	1.85
6000	3.279	2.296	3.74	1.43

Coclusions

A simple but effective system using commercial alumuna mesoporous membrane has been designed to measure and compare the permeation of single gases (H_2 and N_2) at 0.05 to 1.0 bar feed pressure and 298 K. Higher flow rates were obtained for H_2 and N_2. The ratio of H_2 and N_2 flow rates used to calculate H_2/N_2 selectivity.

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