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An initial investigation of a nano-composite silica ceramic membrane for hydrogen gas separation and purification

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ABSTRACT

The effects of trans-membrane pressure difference on hydrogen recovery are investigated on a preliminary basis. The membrane has been developed for high temperature hydrogen separation from CH₄, N₂, and Ar gas molecules. Gas permeation and separation performance of the developed membrane was evaluated. The silica fabricated membrane exhibited high H₂ gas flux and much lower fluxes for CH₄, N₂, and Ar and showed a rise in permeance with the inverse of the square root of temperature for H₂. Plots obtained with respect to the effects of gauge pressure on H₂, N₂, CH₄ and Ar component gas fluxes through the modified support (silica membrane) at a pressure of 1 bar showed that H₂ recorded an almost four-fold high flux value of 0.76 mol/m².s compared to that of 0.33 mol/m².s (CH₄), 0.25 mol/m².s (N₂) and 0.22 mol/m².s (Ar). These results were nearly stable for the temperature range investigated (298 K, 373 K and 473 K). In addition, hydrogen maintained a relatively high permeance value of 1.62 × 10⁻⁵ mol m⁻² s⁻¹ Pa⁻¹ at a low pressure of 0.1 bar under room temperature (298 K) compared to that of N₂, CH₄ and Ar which were much lower throughout the temperature range studied and in agreement with literature. H₂ separation factor (experimental Knudsen) over CH₄, N₂ and Ar were quite close to the theoretical Knudsen and can therefore be experimentally applied through further optimization in the separation of H₂ as a key constituent energy for the future.

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Introduction

Hydrogen as an alternative to fossil fuel can pave the way in the fabrication of various operational energy generation devices including the fuel cell [1]. Consequently high-efficiency fuel cell operation from clean hydrogen can be utilized in all

energy sectors including application in transportation and distributed power. Hence, fuel cell with clean hydrogen-rich fuel is top among solutions sought for in the transitional process to a CO₂ free emission economy and a pathway to sustainable energy in the nearest future. Production of a cost efficient and sustainable clean hydrogen is among the top challenges which must be surmounted for the evolution from

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carbon based (fossil fuel) energy economy to hydrogen based economy [2]. Achieving this level of sustainable energy security and economic performance with current technologies has yielded little result in terms of balancing CO₂ emission abatement and the economic growth [3]. Hence the utilization of an affordable technology which will be readily available and can provide economic and clean source of energy can be set out to meet these challenges. An efficient and cost efficient technology for hydrogen/gas separation is highly required that will have a beneficial effect on the overall expenditure of the entire system. Membrane technology at the moment is being highly applied widely in hydrogen separation because of its simplicity of operation, low energy consumption, option for uninterrupted process and known cost efficiency. In particular, inorganic ceramic membranes have shown evidence of chemical and physical properties, high temperature stability as well as unresponsiveness to sarcastic environment, homogenous pore structure and substantial tensile strength which will conquer the limitations of polymeric membranes [4,5,1]. Due to their inherent features, alumina-based ceramic membranes can easily separate small gas molecule like hydrogen. With their gas permeation values larger than polymeric membranes, these kind of membranes have huge prospect for applications in chemical, petrochemical and energy industry where hydrogen separation in severe conditions results to increased productivity as well as process efficiency. Consequently, porous alumina-based ceramic membranes covered with a thin selective layer manufactured by chemical vapour deposition (CVD) or sol–gel methods have attracted great attention for hydrogen separation [6–8]. According to most literature [9–12], silica membranes prepared by different methods like CVD or sol–gel, deposited on mesoporous or macroporous supports have been shown to be effective for H₂ permeation with good selectivities. In their work, Oyama and Lee [11] and [12] used chemical vapour deposition method to deposit silica on γ -alumina supports with pore diameter of 4 nm and obtained silica membranes with high selectivities of hydrogen for temperatures less than 900 K. Yan et al. [13] used chemical vapour deposition to add silica layer to a macroporous α -alumina support which had 110–180 nm pore diameter with three layers of γ -alumina. They obtained a membrane with selectivity for H₂/N₂ of 100–1000 and a H₂ permeance of 10^{-8} – 10^{-9} mol m⁻² s⁻¹ Pa⁻¹ at 873 K. Oyama et al. [14] reported that preparing a composite layer by silica and an inorganic oxide like zirconia (ZrO₂) gives rise to membrane stability with superior permeation properties. Nomura et al. [15] found an improvement of steam stability of a silica membrane. Although this membrane had a good selectivity for H₂/N₂ of over 800 at 773 K, the permeance was low and in the order of 2 – 7×10^{-8} mol m⁻² s⁻¹ Pa⁻¹. In another work [14], Lee et al. [16] prepared membranes with silica layer on the outer surface of a mesoporous alumina support and obtained a H₂ permeance of 1.2×10^{-7} mol m⁻² s⁻¹ Pa⁻¹ and a selectivity of H₂ to CH₄ of 2800 at 873 K. Gu and Oyama [17], and [18] effectively produced a defect-free γ -alumina multilayer membrane with an ordered structure by successively placing boehmite sols of different particle sizes on a macroporous alumina support. They now coated the surface of the γ -alumina with a thin-silica by chemical vapour deposition method. The resulting novel silica-on-alumina membranes

had excellent permeability of 5×10^{-7} mol m⁻² s⁻¹ Pa⁻¹ and good selectivity for hydrogen over CH₄ of 1500–5900 at 873 K. The resulting supported composite silica–alumina membrane has high permeability for hydrogen in the order of 10^{-7} mol m⁻² s⁻¹ Pa⁻¹ at 873 K with a selectivity of H₂ over CH₄ of 940. However, silica membrane fabrication using similar methods as stated above by deposition on support of large pore (macro porous) are known for their effectiveness with excellent H₂ permeation selectivity outcomes [19,20,11,12]. Again, the difference in the flow rate and permeation features during the separation processes involving multiple component gases makes silica ceramic membrane a good contender for H₂ gas separation [21]. However the main features in the selection of silica based membranes are their hydrogen separation ability and large hydrogen permeability [17].

In this work, a preliminary experiment has been performed using a novel nano-porous silica ceramic membrane manufactured through another alternative, dip coating method. The membrane separates hydrogen gas from N₂, Ar, and CH₄. In particular, the membrane can be applied to separate hydrogen at low-pressure from gas streams and also at elevated temperatures. The process of preparation of the membrane and its description, pure gas fluxes and permeance through the membrane, as well as its industrial application are reported. The morphology of the silica membrane layer is analysed by scanning electron microscopy. However, the transport properties of the membranes were obtained in the temperatures of 298 K, 373 K and 473 K and at pressure differences ranging from 0.1 to 1 bar.

Experimental methods

Preparation of a nano-composite ceramic membrane

The nano-composite membrane in this work was prepared by depositing a very thin, dense layer composed of silica based solution on a macro-porous alumina support for the purpose of modification. Fig. 1 shows the schematic diagram of the sequential steps that are employed to carry out synthesis and characterization of a membrane before permeation tests takes place.

The commercial alumina (SCT, France) tube, (i.d. = 7 mm, o.d. = 10 mm) with a nominal pore size of 15 nm and surface area of 0.0062 m² was used as the support and has been modified by dip coating technique. This dipping process used a silica-based solution comprising of 900 mls of 2-methyl-butane, 100 mls of silicon elastomer and 10 mls of a curing agent. The mixture of Iso pentane, silicone elastomer and curing agent (which prevents a cross linking between silica molecules) were mixed together thoroughly using a magnetic stirrer for homogeneity purposes. A fresh ceramic support was immersed into the solution prepared and left for about 30 mins; it was then withdrawn and placed on a rotating device for approximately 1 h for air drying. After this, it was put in an oven at a temperature of 65 °C for another 2 h. This process is a patented innovation [22,23]. In addition the entire procedure was done to achieve the desired affinity of the membrane for the gas of interest, in this case hydrogen.

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