Effect of SBA-15 intermediate layer of hydrogen permeation of porous alumina-supported palladium membrane

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Highlights

- SBA-15 as an intermediate layer improves H_2 permeability
- higher hydrogen selectivity is obtained at lower thickness
- The prepared membrane is intact at 2 bar pressure and 400 °C temperature

1. Introduction

Global energy demand is expected to rise due to the rapid growth of emerging economies. Maintaining the current energy system, based on the massive use of fossil fuels, the global warming caused by anthropogenic carbon dioxide emissions will worsen[1]. A gradual transition phase of energy demand will begin to find renewable energy-based fuels for the future. Hence, several countries are pursuing ambitious policies and making major investments to develop new technologies. Furthermore, improvements in energy efficiency and improvement in current processes are expected to account for more than half of the reduction in global carbon emissions in the coming decades. The use of process intensification strategies in the majority of current industrial processes is one of the most challenging alternatives. The hydrogen economy could play a very important role in achieving these transitions. However, it is important to note that hydrogen obtained through thermal processes is always accompanied by other sub-products; therefore, additional separation and purification steps are critical to obtaining hydrogen at the required purity. The use of selective palladium-based dense membranes is an appealing alternative for hydrogen purification. In an ideal scenario, this technology enables 100% H_2 selectivity via a solution-diffusion permeation mechanism while maintaining appropriate permeates fluxes at high temperatures However, this is possible only if the prepared membrane is free from any defects and of lower thickness. Hence, self-supported membranes which are of larger thickness (few mm) are of limited use and supported membranes are preferred. Ceramic support (Alumina support) provides a relatively smooth surface and can sustain high temperature. However, they have wide pore size distribution[2] and hence, though they provide good flux a large palladium layer need to be deposited to achieve the desired purity. To overcome this several authors have tried to deposit some intermediate layer to minimize the metal (Pd) use.

In this work, a SBA-15 layer is deposited as an intermediate layer on porous alumina support to prepare a Pd-Cu membrane for hydrogen separation. The effect of SBA-15 size on effectiveness of intermediate layer is investigated. The prepared membranes are tested at different temperature, pressure and gas composition to find their performance,

2. Methods

The membrane is prepared on a porous alumina support of OD 10mm, ID 7mm and length 150 mm. SBA-15 is prepared by different methods (hydrothermal method, sol-gel) to maintain different size of the SBA-15 particles. The prepared SBA-15 layer is deposited over the porous alumina tube by using vacuum assisted dip coating method. The tube is died at 140 °C for 6 hours and then calcined at 650 °C for 8 hours. Thereafter, Pd-Cu layer is deposited simultaneously by using electroless plating (ELP) method. The annealing of prepared membrane is performed in a specially designed reactor with 10% hydrogen and 90% nitrogen mixture at 550 °C for 5 hours. The prepared membrane is characterized and then tested in a modular membrane separator unit. The effect of temperature, pressure, inlet feed rate and gas composition is studied. The inlet, permeate, and retentate side flow rate and gas composition are analyzed by using flowmeter and gas chromatography respectively for all the conditions. The acquired data is used to calculate the hydrogen selectivity and permeate flux to compare the performance of different membranes.

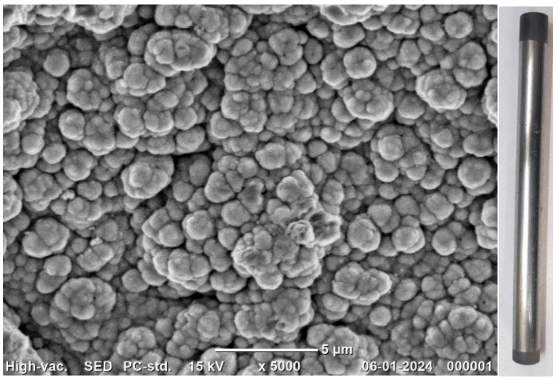

Figure 1. Dense Pd-Cu/Pd loaded membrane SBA-15 and dense membrane

3. Results and discussion

Figure 1 shows the SEM image and photograph of prepared Pd-Cu membrane. The SEM image shows cauliflower structure which is formed due to palladium grains. Further, no pin holes are observed which confirms that membrane is dense. The prepared membrane is further tested in a modular membrane separator at different temperature and pressure. The experiments are performed with pure hydrogen, H_2/N_2 mixture and $H_2/N_2/CO/CO_2$ mixture of different compositions. Figure 2 shows the permeate flux vs ΔP plot for different pressure at 300°C temperature. The results show that for n=0.5 the R^2 value is maximum. This confirms that the synthesize membrane follows the Sievert's law and hence confirms that the prepared membrane is dense. The hydrogen flux of 5.37 x 10^{-4} mol m^{-2} is observed at ΔP = 100 kPa. The H_2/N_2 selectivity more than 1500 are obtained. Similar results will be presented for different temperature, pressure, and feed composition. Further, membrane prepared by using different SBA-15 size will be compared.

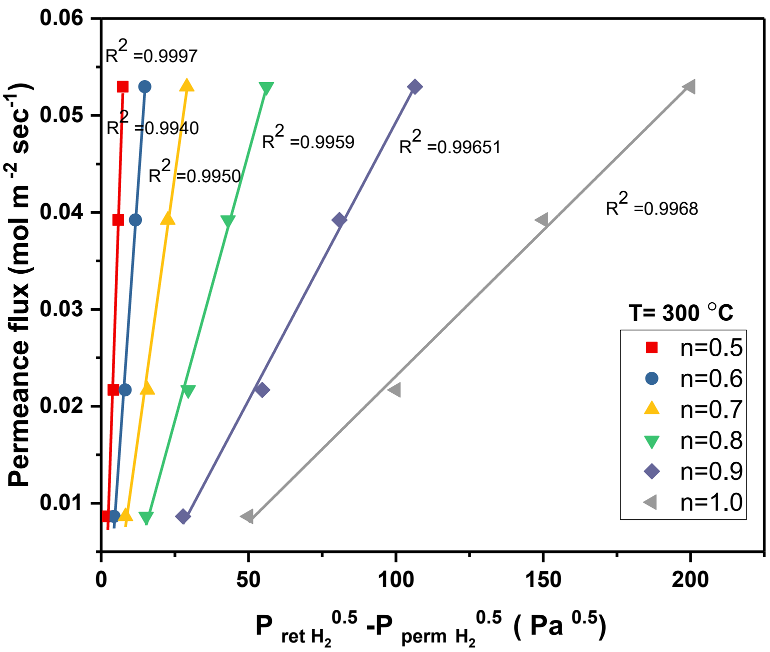

Figure 2. H_2 flux vs transmembrane pressure at 300 °C temperature

4. Conclusions

The current work presents importance of SBA-15 intermediate layer on performance of alumina supported Pd-Cu membrane. The results show that the intermediate layer property have significant effect on membrane densification and hence the performance.

References

[1] Muradov, N. Low to near-zero CO2 production of hydrogen from fossil fuels: Status and perspectives. *International Journal of Hydrogen Energy* **2017**, 42, 14058–14088.

[2] Sharma, R. , Kumar, A. , and Upadhyay, R. K. Catalytic Sol Assisted Dense Pd/γ-Al2O3 Membrane using Modified Electroless Plating: Effect of Process on Surface Morphology. *Chemical Product and Process Modeling* **2017**, 12

Keywords

"Supported membrane, Alumina tube, Surface Modification, Palladium, Pore-Plating Electroless Plating Method.