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Synthesis and performance evaluation of Nanocomposite SAPO-34/ceramic membranes for CO/N mixture separation

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ABSTRACT

Global warming, resulting from emission of greenhouse gases (GHGs), is the cause of drastic climate changes that threatens the economy and living conditions on the planet. Currently, recovery and mitigation of these greenhouse gases remains a technological and scientific challenge. Various recovery processes for the mitigation of GHGs have been reported including among others carbon capture and storage (CCS). The most mature and applied technology in CCS process involves the absorption of carbon dioxide on amine based solvents. However, studies have shown that this process has several drawbacks that include low stability and high energy required to strip off the absorbed CO_2 and regenerate the solvent. This presents an opportunity for the development of new materials for CO_2 capture such as zeolite membranes.

Previous studies have shown that the separation of CO_2 can be achieved with high selectivity at low temperatures using thin-film SAPO-34 membranes (thin layers on supports). This is because CO_2 adsorbs strongly on the membranes compared to other gases found in flue gas. In the thin-film membranes supported on ceramic or sintered stainless steel, thermal expansion mismatch may occur at higher operating temperatures resulting in loss of membrane selectivity due to the formation of cracks. A new method is required to overcome the aforementioned problems, thereby enhancing the separation application of the membranes at higher temperatures.

The effective separation and capture of CO_2 from the coal-fired power plant flue gas is an essential part in the CCS process (Figueroa et al., 2016; Yang et al., 2008). Currently, the capture stage is a huge contributor to the overall cost of CCS (Yang et al., 2008). This is due to the high-energy intensity and inefficient thermal processes applied in the separation and capture in various industrial applications (Yang et al., 2008).

This work presents the use of nanocomposite SAPO-34 zeolite membranes synthesized via the pore-plugging hydrothermal method for the separation of CO_2 during post-combustion CO_2 capture. The SAPO-34 membranes used were supported on asymmetric α-alumina as membrane supports. The membranes were characterized with a combination of dynamic and static physicochemical techniques such as Basic Desorption Quality Test (BDQT), X-ray diffraction (XRD) spectroscopy, Scanning Electron Microscopy (SEM), Fourier Transform Infrared (FTIR) spectroscopy and thermogravimetric analysis (TGA). The characteristic peaks

at 2θ = 21°, 26°, and 32° on the XRD pattern confirmed the presence of SAPO-34 with a rhombohedral crystalline structure. The SEM images showed the formation of the cubic crystalline which were consistent with the reported morphology of SAPO-34. FTIR spectra showed the presence of the essential double-6 membered rings (D6R) and TO_4 structural groups in surface chemistry of crystalline materials further confirming the presence SAPO-34. The TGA confirmed that the membranes possessed high thermal stability.

To assess the feasibility of the synthesis process, the nanocomposite zeolites were grown within the tubular supports. The SEM images of the cross-section of the membrane confirmed the presence of the zeolites within the pores of the support confirming the fabrication of nanocomposite membranes by the pore-plugging synthesis method. The permeation tests used a dead-end filtration mode to measure the single gas permeance and the ideal selectivity of CO_2 and N_2 were calculated.

The BDQT was essential in the study of the quality of the as-synthesized nanocomposite membranes. The quality of the membranes increased with an increase in the synthesis layers of the membranes. However, with an increase in synthesis layers, the membrane thickness also increases. The membrane thickness affected the gas permeance for CO_2 and N_2 significantly. The permeance of the N_2 gas decreased from 10.73 $x10^{-7}$ $mol.s^{-1}.m^2Pa^{-1}$ after the first synthesis to 0.31 $x10^{-7}$ $mol.s^{-1}.m^2Pa^{-1}$ after seven synthesis layers. Alternatively, the more adsorbing gas CO_2 decreased from 12.85 $x10^{-7}$ $mol.s^{-1}.m^2Pa^{-1}$ to 2.44 $x10^{-7}$ $mol.s^{-1}.m^2Pa^{-1}$. The performance of these zeolite membranes depends significantly on the operating conditions. Hence, we studied extensively the influence of the various operating conditions such as temperature, feed pressure and feed flowrate in this work.

Results indicated that the membrane separation performance in this study is largely dependent on the temperature. In addition, the ideal selectivity decreased significantly with an increase in temperature. High temperatures results in less adsorption of the highly adsorbing CO_2 gas, the permeance reduces significantly, while the permeance of the less adsorbing N_2 increased slightly. The feed flow rate has less effect on the adsorbing gas while the non-absorbing gas increased resulting in a decrease in the ideal selectivity as well. The nanocomposite membranes in this study have a low flux compared to their thin film counterparts. An increase in feed pressure significantly increased the flux significantly as well as the ideal selectivity.

Maxwell-Stefan model simulation was done in this study to describe the permeance of pure CO_2 single gas permeance as a function of temperature. This model considered explicitly the adsorption-diffusion mechanism, which is the transport phenomenon, involved in the transport of CO_2 through the zeolite membrane. The description of the support material was included in the model as well. However, the model was only applied to the CO_2 gas permeation well within the experimental data. We then compared the model was with the experimental results and a good correlation was observed.

In conclusion, SAPO-34 nanocomposite zeolite membranes were obtained at low temperatures (150 °C) with a short synthesis time (6 h). In addition, the high thermal stability of the as-synthesized SAPO-34 membranes makes them ideal for high temperature CO_2 separation such as the intended post-combustion carbon capture. The BDQT revealed that the quality of the membranes was related to the thickness of the membranes. Therefore, better membrane quality was obtained with relatively thicker membranes. The separation performance evaluation was conducted on the membrane with the greatest quality. Our findings demonstrate that the performance of the membranes depends extensively on the operating conditions.