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PERFORMANCE OF ZEOLITIC IMIDAZOLATE FRAMEWORK-8 ON QUARRY DUST-BASED CERAMIC HOLLOW FIBRE MEMBRANES FOR CO2 AND CH4 PERMEATION

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ABSTRACT

Raw natural gas typically contains undesirable impurities such as CO_2, water and heavy hydrocarbons. As the demand for natural gas increases, an effective way to remove CO_2 from natural gas using a membrane has been proposed. Ceramic membrane has been considered as potential candidates than polymeric membrane for industrial application due to its ability to be used in harsh conditions. However, the fabrication of ceramic hollow fibre membrane (CHFM) has larger footprint due to multiple steps fabrication and expensive raw material that simultaneously increased the overall cost of ceramic membrane applications. Therefore, by using low-cost raw materials, particularly from quarry dust waste, the cost can reduced significantly. Up until no study have investigated the used of granite dust for CHFM fabrication. In order to further enhance the selectivity of ceramic membrane, zeolitic imidazolate framework-8 (ZIF-8) layer was deposited onto the ceramic hollow fibre membrane (CHFM) support. Pebax was also added to improve the compatibility of ZIF-8 with CHFM and further enhance the CO_2 permeability. The pyscochemical properties of quarry dust was first characterized using XRD, XRF, BET, TGA, FTIR, and particle sizer before being used as the raw material for CHFM fabircation. The CHFM was prepared via a phase inversion and sintering method where the effects of bore fluid flow rate (6-10 $ml \cdot min^{-1}$) and sintering temperature (1050-1150 °C) towards the morphology and microstructure, pore size distribution, pore volume and porosity, mechanical properties and pure water flux were then systematically investigated. Higher sintering temperature enhances the mechanical properties of CHFMs due to pores densification, but it also leads to a less porous CHFM, which can affect their permeation flux. The relatively low sintering temperature needed for QD-based CHFM (<1150 °C) offers a reduction in energy consumption, which is economically attractive for future commercialization of CHFM. In order to address the second objective, different loading of ZIF-8 (0.4-1.6 wt%) was added in Pebax solution before being deposited onto QD-CHFMs. The effects of ZIF-8 coating layer on the physicochemical properties and morphological structure of the membranes were them investigated. By controlling the ZIF-8 loading, a uniform and well-coated ZIF-8 selective layer with different thickness was formed on the QD-CHFM. A single gas permeation study for CO_2 and CH_4 gasses was then conducted at 2-6 bar. It was observed that the permeability of CO_2 gasses were higher , making it suitable for future CO_2 separation membrane application. The use of lower loading of ZIF-8 leads to better permeability ($1.19x\ 10^{-4}\ cm^3(STP) \cdot cm \cdot cm^{-2} \cdot s^{-1} \cdot cm^{-1} \cdot Hg^{-1}$) and selectivity (4.19) as compared to higher loading of ZIF-8. This was because the increased of ZIF-8 loading in the Pebax coating solution caused agglomeration of ZIF-8, which simultaneously increased of pathway tortuosity. Therefore, the gas diffusion rate and permeability across the membrane decreased. The performances of ZIF-8/QD-CHFM were observed to be at par compared to other ceramic membranes available in the literature. In this work, QD waste successfully recycled as a raw materials for CHFM fabrication, which helps to minimize the disposal issue of quarry dust and environmental pollution. At the same time, the overall cost of CHFM can be reduced for future commercialization. The deposition of ZIF-8 membrane can enhance the overall properties of the membrane. However, the loading should be appropriate controlled as it can significantly affect the gas separation performances.

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CHAPTER ONE
INTRODUCTION

1.1 Research Background

Natural gas is one of the most efficient energy sources and the demand for natural gas is increasing rapidly yearly. However, other than methane (CH_4), some other inorganic gases are also present such as carbon dioxide (CO_2), helium (He), water vapor (H_2O) and hydrogen sulphide (H_2S). Removal of CO_2 from natural gas is essential to prevent corrosion on pipelines and equipment. Various processes of CO_2 removal have been widely utilized over the years such as absorption, adsorption process and cryogenic distillation. However, some of these conventional techniques require a gas-to-liquid phase change, additional solvent, and chemicals for separation, which adds a significant energy and operating cost to the separation cost.

Gas separation over a membrane has been receiving great attention for CO_2 removal recently due to the simplicity in the process and compact unit. In addition, it is more environmentally friendly and does not require phase changes, leading to low capital investment costs and low energy utilization [1]. A membrane is a thin barrier between two phases or mediums that allows one or more constituents to selectively pass from one medium to another while retaining the rest. The separation occurs with the presence of an appropriate driving force such as concentration, temperature, pressure, or electrical gradients. For gas separation application, the membrane is typically a non-porous layer. The selective gas separation can be obtained by appropriately tuning the selected species' permeability using a specific material design.