

Synthesis and performance evaluation of optimized silica sodalite in polysulfone mixed matrix membranes for pre-combustion CO₂ capture

C Eden (803 769)

Supervised by M.O. Daramola

School of Chemical and Metallurgical Engineering, Faculty of Engineering and the Built Environment, University of the Witwatersrand, Johannesburg, South Africa, for the Degree of Master of Science in Chemical Engineering

Abstract

Increasing carbon dioxide emissions, resulting primarily from increasing energy demand and industrial activity, are a major concern for environmental climate change. This has led to a global movement towards CO₂ capture from point source emitters such as power plants. Membranes designed to purify H₂ from CO₂ and CO, such as in pre-combustion coal gasification, need to be thermally and mechanically stable with good separation performance in order to make them an economically attractive alternative to traditional separation methods. Polymeric membranes, although mechanically stable and inexpensive, exhibit a trade-off between permeability and selectivity. This limits their separation performance. Mixed matrix membranes which combine polymer matrix with inorganic zeolite particle, have the potential to increase the separation performance of H₂ from CO₂. Hydroxy sodalite (HSOD) is a common zeolite with potential applications in absorption and catalysis. The small cage aperture enables the permeation of small molecules such as hydrogen. However, traditional hydrothermal synthesis of HSOD results in occluded water and organic matter within the cage structures, hindering permeation. Attempts to remove these occluded molecules, through dehydration above 400 °C, result in the collapse of the cages, rendering the substance completely ineffective. Topotactic conversion has the potential to produce higher quality sodalite without the presence of occluded matter. However, previous research has exhibited discrepancies in the quality of the sodalite synthesized and the synthesis variables utilized.

This study aimed to produce high porosity silica sodalite through topotactic conversion and investigate the effects of synthesis variables such as; acid strength, acid treatment time, and

calcination time and temperature on the textural and morphological quality of the sodalite. Silica sodalite was synthesized from the fabricated RUB-15 precursor and converted through topotactic conversion to SSOD through treatment with propionic acid and calcination. The synthesized sodalite was then utilized in mixed matrix membranes (silica sodalite/ polysulfone (PSF)) by dissolving the various loadings in PSF solution and casting to a thin film. The membranes were tested for hydrogen/carbon dioxide separation potential, membrane strength and thermal stability.

Preliminary investigation of the effect of synthesis variables via the response surface modelling approach revealed acid treatment time to be an insignificant influencing factor affecting the quality of silica sodalite prepared via topotactic conversion and was thus removed from further investigations. Consequently, higher acid strength, lower calcination time and temperature were observed to favour higher quality silica sodalite. Therefore, optimum synthesis variables obtained via the RSM approach were acid strength: 5 M, calcination time: 1 h treatment and calcination temperature: 873 K. The sodalite reproduced under these conditions displayed higher surface area and pore volume than the HSOD crystals produced via traditional hydrothermal synthesis of 2.35 m²/g and 0.012 cm³/g, respectively but fell short of the model predicted SA and PV values of 148.5 m²/g and 0.093 cm³/g, respectively.

The subsequent stage of the study considered the utilization of the silica sodalite crystals as a filler in PSF membranes to form SSOD/PSF mixed matrix membranes. The improved separation performance of the fabricated membranes over that of the pure PSF membrane was evaluated via single gas permeation and mixed gas separation using pure H₂ and CO₂; and mixed CO₂/H₂ gas (40:60), respectively. The tests were carried out using a custom-built gas separation set-up at atmospheric temperature (25 °C) and pressures from 1-5 Bar. The addition of the filler lead to improved permeability (582 % > PSF) and permeance (184 % > PSF) over the pure PSF membrane, at the cost of reduced H₂/CO₂ selectivity (68 % < PSF). Increasing the filler loading produced a parabolic effect, suggesting a cap on permeability enhancement at 4.246×10^{-11} mol.m/m².s.Pa in the 10 wt. % membrane through the addition of silica sodalite. The results of this study demonstrate the enhanced separation potential of silica sodalite filled mixed matrix membranes for gaseous applications if defect free membranes can be fabricated.