Abstract

It is generally accepted by the scientific community that CO_2 is a major greenhouse gas responsible for global warming, marked by an increase in global temperature. Amongst other sequestration options to minimize the impact of CO_2 on the environment, carbon capture and storage (CCS) is essential. The technology already implemented and validated for separating CO_2 from other gases on an industrial scale is the absorption technology using aqueous amine. However, the use of amine scrubbing is a costly operation. Membrane technology is one of the best separation processes adopted for liquid separation, a cost-effective method and is considered for gas separations as the technology is expected to deliver lower energy penalties.

Zeolite-based membranes that can withstand aggressive chemicals, high temperatures and pressure operations are very appealing for use in pre-combustion CO_2 capture. However, zeolite membranes synthesized with zeolite crystals coated on the surface of the support, otherwise known as "thin-film" supported membranes are prone to defects. This leads to loss of selectivity of the membrane. Moreover, the formation of cracks is promoted when thin-film zeolite membranes are exposed to high temperatures, which occur as a result of the huge difference in thermal expansion of the supports in comparison to the zeolite crystals. Nanocomposite zeolite membrane prepared by embedding zeolite crystals within the pores of the support, yields remarkably thermally and mechanically stable nanocomposite membranes.

In this study, sodalite crystals were prepared via the pore-plugging hydrothermal (PPH) synthesis protocols. After evaluating the sodalite crystals, the PPH protocols that yielded high-quality sodalite crystals were then utilized for the synthesis of high-quality nanocomposite sodalite membranes supported on α-Al_2O_3 ceramic tubes. The as-synthesized sodalite crystals were evaluated using techniques such as scanning electron microscopy (SEM) for checking the surface morphology of the as-synthesized sodalite crystals. X-ray diffraction (XRD) was used to check the crystallinity and purity of sodalite crystals, while Fourier transform infrared spectroscopy (FTIR) was used to evaluate the surface chemistry of the as-synthesized sodalite crystals. The quality of the as-synthesized membranes was evaluated using single gas permeation (H_2, CO_2, N_2) and for separating H_2 from H_2/CO_2 in the context of pre-combustion CO_2 capture.

The main findings of the research revealed that the quality of the as-synthesized sodalite crystals can be positively or negatively impacted by adjusting PPH synthesis conditions such as ageing, pre-interruption and post-interruption time. Furthermore, during single gas permeation test of membranes prepared by the best PPH synthesis protocols compared to membranes synthesized via the direct hydrothermal synthesis method, membrane prepared via PPH synthesis using two interruption steps displayed the best performance. The nanocomposite membrane displayed H_2 permeance of 7.97×10^{-7} mol.s^{-1}.m^{-2}.Pa^{-1} at 100 °C and a feed pressure of 0.48 MPa. Consequently, the H_2/CO_2 ideal selectivity at these conditions was 8.76. Regarding H_2/CO_2 mixture, the permeance of H_2 reduced from 8.03×10^{-7} mol.s^{-1}.m^{-2}.Pa^{-1} to 1.06×10^{-7} mol.s^{-1}.m^{-2}.Pa^{-1} when tested at 25 °C and a feed pressure of 0.18 MPa. Also, in the presence of CO_2, nanocomposite sodalite membrane selectivity (i.e. separation factor) reduced to 4.24 at 25 °C and a feed pressure of 0.48 MPa.

Even though adjusting PPH synthesis protocol of sodalite crystal preparation before synthesis of sodalite membrane produced membrane with interesting results, optimizing the PPH synthesis in the presence of the support might yield better results as the support could play a role during sodalite crystal growth to reduce defect formation. The encouraging results as documented in this dissertation provide a platform for further investigation on sodalite/ceramic nanocomposite membrane for efficient pre-combustion CO_2 capture.