

論文の要旨

題目 PREPARATION OF ORGANOSILICA MEMBRANE AND APPLICATION TO USE IN GAS

SEPARATION AND REVERSE OSMOSIS

(オルガノシリカ膜の開発と気体分離および逆浸透への応用)

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The overall objective of this dissertation research is to study on the preparation of organosilica membrane and application to use in gas separation (GS) and reverse osmosis (RO) applications. The brief descriptions of each chapter in this dissertation are shown below:

Chapter 1 is **“General introduction”** which provides the background and motivation of the current research.

Chapter 2 is **“Effect of firing temperature on the structural and gas separation performance of 2,4,6-tris- [3(triethoxysilyl)- 1-propoxy]-1,3,5-triazine (TTESPT)”** which describes a development on the 2,4,6-Tris[3(triethoxysilyl)-1-propoxy]-1,3,5-triazine (TTESPT) membrane. Since, TTESPT is a new existence organosilica; hence we emphasized more extra study on its potential in gas separation area. The TTESPT-derived silica membrane exhibits a significant degree of selectivity for $C_3H_6-C_3H_8$ ~37 at a permeation temperature of 50 °C, which greatly surpasses the upper-bounds of selectivity and permeance trade-off of carbon membranes. This indicates the potential for further development toward $C_3H_6-C_3H_8$ separation applications.

Chapter 3 is **“Effect of water ratio on the structural and performance evaluation; gas separation and reverse osmosis of 2,4,6-tris- [3(triethoxysilyl)- 1-propoxy]-1,3,5-triazine (TTESPT)”**. In this research, TTESPT membrane has been developed for molecular separation processes in gas (gas separation) and liquid phases (reverse osmosis (RO)). By adjusting the $H_2O/TTESPT$ molar ratio, we found a promising technique for tuning the pore network of TTESPT membranes. A TTESPT membrane with a high $H_2O/TTESPT$ molar ratio exhibited a high degree of selectivity for H_2/SF_6 (greater than 4000) at a permeation temperature of 200 °C and demonstrated high sodium chloride (NaCl) rejection (>98.5%) with water permeability of $>1 \times 10^{-12} m^3 m^{-2} s^{-1} Pa^{-1}$ under operating conditions of 1 MPa and 60 °C during a RO experiment.

Chapter 4 is **“Robust BTESE membranes for high temperature reverse osmosis (RO) application: Membrane preparation, separation characteristics of solutes and membrane regeneration.”** In this study, a pore network of bis(triethoxysilyl)ethane (BTESE) organosilica membranes was controlled by adjusting the molar ratios of $BTESE/H_2O/acid=1/x/0.2$ ($x=3$ and 240). The mechanisms for solute transport in the BTESE

RO membrane were investigated using three different aqueous solutions of sodium chloride (NaCl), ethanol (EtOH) and isopropanol (IPA). It was noteworthy that the rejection of alcohols decreased with an increase in the RO operating temperature, while the electrolyte rejection remained almost constant. The BTESE membranes exhibited high thermal robustness under the long-term testing conditions, delivering salt rejections >98% until the end of the testing period (50 h). The BTESE membranes could also be regenerated after use in the gas and RO experiments, thus demonstrating robust properties.

Chapter 5 is “Favorable of BTESE membranes for gas and reverse osmosis (RO) applications: Effect of preparation conditions on the structural and the correlation between gas and liquid permeation properties.” The aim of this work was to study the simultaneous effects of H₂O/BTESE molar ratio (WR) = (3, 60, 120, 240) in different firing environments on the structural and permeation properties of BTESE membranes. We found that most of organic peaks for sample fired in N₂ environment were more intense as compared with the samples fired in air environment. Thus, the samples fired in N₂ environment are more hydrophobic compared with the sample fired in air environment and can be proven by contact angle and H₂O adsorption results. In term of separation performance, the permeance of gases and H₂O were clearly dependent on WR. Increasing WR decreased the permeance of both gases and H₂O via the pore network of BTESE membrane. On the other hand, changing the firing environments also affected the permeance of gases and H₂O. Samples fired in air environment exhibited higher permeance of gases and H₂O due to more open pore networks. In addition, the relationship between gas and liquid permeances was correlated by assuming He gas as a predictor of water permeance, N₂ gas as a predictor for IPA and SF₆ gas as a predictor for NaCl permeance. Increasing in He and permeance ratio resulted in increasing in water permeance and rejection of solutes.

Chapter 6 is “New insights on fouling and cleaning properties of reverse osmosis (RO) BTESE membrane.”Initially, our attention is restricted to BTESE membranes and to understand the fundamentals of fouling phenomenon as well as its cleaning process. Four types of foulants such as bovine serum albumin (BSA), sodium alginate (SA), sodium dodecyl sulfate (SDS) and dodecyltrimethyl ammonium bromide (DTAB) were chosen. More pronounced fouling was seen for DTAB foulant compared with other foulants. It is worthwhile to point out that inorganic membranes have inherently versatile characteristics; high thermal stability and chemical resistance. Thus, these advantages make them suitable candidates for hot water cleaning conditions. Our main concern is to avoid any types of chemical cleaning agents in order to cleaning BTESE membranes. It was found out the optimize cleaning condition of fouled BTESE membrane by DTAB was by using hot water at 80 °C for 30 minutes.

Chapter 7 is “Conclusions and Recommendations”. Main conclusions and recommendations were presented here.