論文の要旨

題 目 Development of polymer-supported organosilica layered-hybrid membranes and their applications to molecular separation

(高分子支持体を用いたオルガノシリカ積層ハイブリッド膜の開発とその分子分離への応用)

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The development of advanced separation membranes with excellent separation performances, a simple fabrication process and low-cost is always being pursued by membrane developers. Hybrid organosilica membranes are proved to possess high hydrothermal stability, adjustable pore size of silica networks and a superior molecular sieving ability. However, all of these organosilica membranes were fabricated on flat or tubular ceramic supports, applications of them are limited due to the high cost of ceramic support materials and complex fabrication processes, resulting in the difficulties in the scale-up. The objectives of this work are to develop the polymer-supported organosilica layered-hybrid membranes, which are expected to be as good as or even better separation performance than that of previous ceramic-supported organosilica membranes. The main works of this study is as follow:

Chapter 1 is "General introduction". The membrane separation technologies and pervaporation (PV) and vapor permeation (VP) processes were overviewed. Conventional membranes to the dehydration of organic aqueous mixtures and typical structure of different types of membranes were summarized. A novel fabrication of polymer-supported organosilica layered-hybrid membrane was proposed, and these membranes were applied to dehydration of organic aqueous solutions in vapor permeation.

Chapter 2 is "Sol-gel spin coating process to fabricate a new type of uniform and thin organosilica coating on polysulfone films". Using 1,2-bis(triethoxysilyl)ethane (BTESE) as the precursor and 1-propanol as the solvent, a uniform and defect-free hybrid organosilica thin coating was successfully deposited onto non-porous polysulfone (PSF) thin films via a sol–gel process using spin coating. The preparation conditions, the structural characterizations, morphology and surface roughness of the BTESE-derived silica coating on PSF film were investigated. Results showed that the outermost layer of the composite film consisted of nanoscale hybrid organosilica networks that were approximately 200 nm in thickness with a low surface roughness.

Chapter 3 is "Fabrication of a layered hybrid membrane using an organosilica separation layer on porous polysulfone ultrafiltration supports, and the application to vapor permeation". Using BTESE as a single precursor, a perm-selective organosilica separation layer was successfully deposited onto porous polysulfone ultrafiltration (PSF-UF) supports via a simple sol-gel spin-coating and thermal-treatment process. The layered hybrid membranes, where BTESE-derived SiO_2 is deposited on porous polymer supports, were applied to the vapor permeation dehydration of isopropanol/water (90/10 wt%) solutions at 105 ºC, and demonstrated a water flux of 1.6 kg/(m^2 h) and a separation factor of 315 with no selectivity for a PSF-UF support. Long-term stability testing of vapor permeation also confirmed the excellent stability of these layered hybrid membranes.

Chapter 4 is "Improving separation performance of layered hybrid membrane by fabricating thin organosilica layer on polymeric nanofiltration membranes". In order to improving separation performance of layered hybrid membrane, the BTESE separation layer was deposited onto the polymeric nanofiltration membranes. The optimal preparation conditions of this membrane were firstly investigated. Then, the membranes were employed to the vapor permeation dehydration of isopropanol/water solutions, and showed a stable water flux of 2.3 kg/(m^2 h) and an improved separation factor of about 2500, which increased to approximately 5-fold compared with pristine polymeric nanofiltration membranes. In addition, single-gas permeance through this membrane was also discussed and a modest H_2/N_2 selectivity of 26 was obtained, which approximates those of ceramic-supported BTESE-derived silica membrane. Finally, these membranes were also tried to apply to reverse osmosis (RO) desalination experiment and good RO performance was also confirmed.

Chapter 5 is "Conclusions". Several important conclusions of this study are given in detail and suggestions are provided for further study.