学位論文全文の要約

Title　Control of separation functional layer structure of polyamide membrane for improved water permeability

（高透水化のためのポリアミド膜の分離機能層構造制御）

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The objectives of this work are to prepare polyamide membranes of high water permeability, good solute rejection and to study separation/permeation mechanism and polyamide formation mechanism. The main work of this study is as follows.

1. Preparation and evaluation of multi-layered polyamide membranes with high water permeability by optimizing the types of added co-solvent into the organic phase.
2. Preparation and evaluation of multi-layered polyamide membranes with high water permeability by controlling the concentrations of added co-solvent into the organic phase.
3. Scaled-up trial and evaluation of module by using mass-production machine.
4. Application of organic-inorganic nanocomposite technology to separation membrane.

To describe the topics and contents more clearly, the thesis is broken down into six chapters. These chapters are briefly outlined as follows.

We proposed a novel fabrication method of polyamide membrane by adding organic solvent into the organic phase during interfacial polymerization of 1,3-phenylenediamine (MPD) with 1,3,5-benzenetricarbonyl trichloride (TMC) on polysulfone ultrafiltration supports. In a novel polymerization method, co-solvents, which included acetone, ethyl acetate, diethyl ether, toluene, isopropyl alcohol (IPA) and N,N'-dimethyl formamide (DMF), were added into the organic phase which made it possible to control the surface morphology and polyamide network structures. As-prepared membranes showed multi-layered ridge-and-valley structures, and the types of co-solvent successfully controlled water permeability and rejection. Polyamide membranes prepared by the addition of 2 wt% ethyl acetate, showed the best performance with NaCl rejection of 99% and water permeability of more than 1 $m^3/(m^2\ d)$ at 1.5 MPa, which was about twice that of a membrane prepared without a co-solvent.

The as-prepared membranes also could control both permeate flux and rejection reactions successfully by the concentration of co-solvents that were added. The optimal membrane conditions included the addition of 3 wt% ethyl acetate, with a NaCl rejection of more than 99% and a permeate flux of more than 1.8 $m^3/(m^2\ d)$ at 1.5 MPa, which was more than three times higher than the membranes prepared without a co-solvent.

Approximately 1m widths flat sheet novel RO membranes which had high values for both salt rejection and permeate flux, were manufactured with the addition of various concentrations of ethyl acetate into the organic phase using a mass production machine and applied to 1m length spiral wound modules 2-inch in diameter. These membranes also showed a specific surface morphology. Furthermore, we found that salt rejection and permeate flux could be controlled widely by changing the concentrations of ethyl acetate. The optimal membrane module prepared by adding 1 wt% ethyl acetate, showed $MgSO_4$ rejection of more than 99.6% and a water permeability of more than 4 m^3/d at 1.0 MPa, which was more than 2-times higher permeate flux than that of the commercial NF module. Furthermore, these membranes showed higher rejection for several kinds of pharmaceuticals than that of the commercial NF membrane.

We also proposed a novel method for fabrication of the polyamide/inorganic particles nanocomposite membranes by using interfacial polymerization technique. These membranes fabricated by interfacial polymerization consisting of PA/zeolite composite functional layer showed high values for both salt rejection and water permeability. Furthermore, we found that salt rejection and water permeability could be controlled by the concentration and the sizes of added zeolite particles. Permeate flux and rejection of as-prepared PA/zeolite nanocomposite membrane were successfully controlled by the concentrations of zeolite that were added. The optimal conditions included the addition of 0.38 wt% zeolite, with a NaCl rejection of more than 98.5%, a $MgSO_4$ rejection of more than 99.5% and a permeate flux of more than 1.1 $m^3/(m^2\ d)$ at 1.5 MPa, which was more than twice permeate flux than that of the membrane without adding zeolites.