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# NOVEL THIN-FILM COMPOSITE POLYAMIDE MEMBRANES PREPARED BY INTERFACIAL POLYMERIZATION FOR PERVAPORATION SEPARATION OF WATER FROM ETHANOL

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## ABSTRACT

To improve the pervaporation performance of polyamide membrane, novel thin-film composite (TFC) polyamide membranes were prepared by interfacial polymerization of novel water-soluble 2,2'-dimethylbenzidine hydrodiamine (m-tolidine-H) and toluene-soluble trimesoyl chloride (TMC) on the surfaces of modified asymmetric polyacrylonitrile (mPAN) membrane. Effects of aqueous amine solution immersion time and polymerization time on the pervaporation performance through m-tolidone-H-TMC/mPAN composite membranes were investigated. The permeation rate decreases with increasing both aqueous amine solution immersion time and polymerization time for pervaporation separation of 90wt% ethanol water mixture through the m-tolidone-H-TMC/mPAN membrane. The polyamide/mPAN composite membrane prepared by immersing 1.5wt% aqueous m-tolidine-H solution for 10s and then contacting with 0.05wt% TMC for 10s has the best pervaporation performance. The water concentration in permeate approaches 99wt% and a 2300 g/m<sup>2</sup>h permeation rate with a 90 wt% aqueous ethanol solution at 25°C was obtained.

## INTRODUCTION

Due to the high thermal stability, excellent mechanical strength and high resistance to organic solvents polyamide is often used to be prepared as polymer membrane for liquid separation process. Although polyamide membrane has high salt rejection in water treatment process because of its rigid and dense structure, but the flux through the membrane is too low. For this reason, the morphology of membrane should be converted from a dense structure to asymmetric or composite type structure.

Thin-film composite membrane is usually obtained by forming an ultra-thin dense layer on a porous substrate through interfacial polymerization. Polymerization reaction takes place at the interface of the two liquids which are insoluble to each other. The skin layer plays a key role in the process of molecular diffusion and separation functions in separation process.

The thin-film composite membranes are usually studied in reverse osmosis or nanofiltration, but there have been a few reports on pervaporation. In this study, novel amine, 2,2'-dimethylbenzidine hydrodiamine (m-tolidine-H), and acyl chloride, trimesoyl chloride (TMC), were used as the monomers of the polyamide active layers prepared on the modified polyacrylonitrile (mPAN) support membranes through the interfacial polymerization. Effect of aqueous amine solution immersion time and polymerization time on the pervaporation separation of 90 wt% aqueous ethanol solution was investigated. It was expected to have good pervaporation performance.

## EXPERIMENTS

The studied membrane is a composite system with an asymmetric layer structure: a polyester nonwoven base

substrate (ca. 150 μm thick), a porous supporting PAN (polyacrylonitrile ca. 50 μm thick), and the top skin layer of interfacial polymerized polyamide (PA). A chemical structure of monomers of polyamide active layer was shown in Fig. 1.

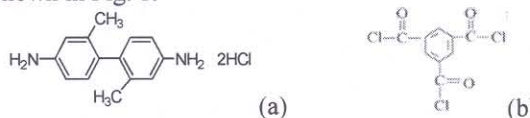


Fig. 1 Chemical structures of monomers used in interfacial polymerization.

The mPAN support surface was immersed in 1.5wt.% aqueous amine solution for a second. Removed excess amount of aqueous amine solution remained on the mPAN support. The mPAN membrane soaked the aqueous amine solution was contact with toluene solution containing 0.05wt.% acyl chloride (TMC) for a while to carry out the interfacial polymerization.

After the removal of the toluene solution, the resulting thin-film composite membranes were stored in methanol overnight in order to remove residual solvent. After that, take out and dried in air. The procedure of interfacial polymerization is shown in Fig. 2.

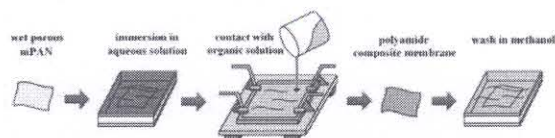


Fig. 2 The procedure of interfacial polymerization.

The polyamide thin-film composite membranes are utilized in the pervaporation for the aqueous ethanol solution.