# 学 位 論 文 内 容 の 要 旨

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## 学 位 論 文 題 名

Membranes Made with Zeolite and ZIF-8, and Their Applications to Water Separation from Organic/Water Mixtures
（ゼオライト膜及び ZIF-8 膜の合成と有機物水混合溶液分離への応用）

A new separation process has been proposed which is instead of the traditional separation by distillation which can consume less amount of energy but achieve a high purity separation. A pervaporation by using membrane is a promising technique to achieve this goal. The membrane process can separate certain molecular mixtures effectively and economically without any toxic or by-production. The materials used for membrane preparation are not limited but cover in a wide range. The structures and properties of the materials decided the membrane performance and the field of membrane applications. The objective of this study is selective separation of water from water/organic mixtures by pervaporation using membrane technology. We studied the method of membrane preparation including the zeolite membrane (mordenite type and MTW type membrane), ZIF materials based membrane (ZIF-8 membrane). We optimized the synthesis method for increasing the performance of the membrane and the membranes were used in the different separation system for application. The whole work is constituted by 7 sections from chapter 1 to chapter 7.
In chapter 2 and 3, the methods of preparation, optimization of the Mordenite membranes and application were discussed, respectively. In chapter 2, Mordenite nanocrystal-layered membranes consisting of a mordenite nanocrystal layer and protection layer were successfully prepared. The effect of nanocrystal layer thickness was discussed to determine the appropriate condition for membrane preparation. The basic conditions were detected in order to use the membrane for pervaporation. The membrane acid stability was examined by separation of water from acetic acid/water solution. In chapter 3, four types of water/organic solvent solutions were prepared for pervaporation experiments using the mordenite nanocrystal-layered membranes to detect the effect of the polarity of the organic solvent in the feed solution on the permeance of water through the mordenite nanocrystal-layered membrane. The mechanism of the mordenite type zeolite membrane was studied and used for directing the membrane preparation. In order to prepare the membrane with high separation ability and permeability, the effects of synthesis conditions on the membranes performance, such as hydrothermal temperature, pre-aging time for the mother liquid which using for hydrothermal synthesis and heating rate during the hydrothermal synthesis were discussed. Moreover, the obtained mordenite membranes were applied to the separation of water from water/organic solutions (organic solvents: ethanol, acetone, 2-propanol, or acetic acid) using a pervaporation method.
In chapter 4 and chapter 5, a new type high-silica material with a unidimensional 12-membered ring channel were used to prepare the membrane. At first, the MTW nanocrystals synthesis with different Si/Al ratios and different kinds of the organic structure directing agent (OSDA) were discussed to dicided the seed crystals for membrane preparation. A MTW-type zeolite nanocrystal-layered membrane composed of nanocrystal and protection layers were successfully prepared by a secondary growth method under hydrothermal conditions. The acidic proof ability of MTW membrane was detected by separation of water from acetic acid/water solution. Since the MTW can be synthesized in a wide range of Si/Al and the crystals morphology can be controlled by using different kind of the OSDA molecules, in chapter 5, the effects of crystal morphology, Si/Al ratio and thickness of MTW zeolite membrane on water/2-propanol separation by pervaporation were discussed, respectively. In order to get better understanding of the membrane preparation and separation, the mechanisms of the

MTW type zeolite membrane were discussed in the chapter. The function of the protection layer and the nanocrystal layer in the membrane separation is studied, respectively.
A new kind of material-zeolite imidazolate frameworks (ZIFs) were focused in chapter 6, ZIFs are a subclass of metal organic frameworks which are composed of tetrahedrally-coordinated transition metal ions connected by organic imidazole linkers. Since the metal-imidazole-metal angle is similar to the 145° Si-O-Si angle in zeolites, ZIFs showed zeolite-like topologies. Moreover, ZIFs possess the advantages of both zeolites and MOFs, such as large surface areas, high crystallinities and exceptional thermal and chemical stabilities. Therefore, the synthesis methods of ZIF-8 crystals and ZIF-8 membranes were discussed, respectively. The nanometer size ZIF-8 crystals were used for ZIF-8 membranes synthesis. In order to prepare the high performance ZIF-8 membrane, the synthesis method was discussed. Moreover, the gas adsorption isotherms of ZIF-8 crystals were investigated to understand the properties of ZIF-8 materials. The permeance of single component (water, ethanol, butanol, benzene, and hexane) through ZIF-8 membrane was detected to investigate the application of ZIF-8 membrane.