# A115 Preparation and permeation properties of Y-type zeolite membrane

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## 1. Introduction

Zeolite membranes are expected to play an important role in the separation of variety of materials used in the chemical and petrochemical industries. The most significant progress was achieved by A-type zeolite membrane in the pervaporation dehydration. In previous studies, FAU membranes have been prepared and used for pervaporation and gas separations.[1)] In this work, NaY zeolite membranes were synthesized in various synthetic conditions.

## 2. Experimental

Membranes were prepared on tubular porous mullite supports and porous tubular stainless steel (SS) supports. Seeding was carried out by rubbed the support with NaY powder. A gel used for the synthesis was prepared by mixing sodium silicate powder ($SiO_2$ 59 wt%, $Na_2O$ 21.5 wt %), $Al(OH)_3$ (Wako, 97 wt%) and NaOH. The mixture was stored at 30 °C for 0-24 h prior to the hydrothermal synthesis. The seeded tubes were then vertically placed into synthesis reactor. Hydrothermal treatment carried out at 90–150 °C for a specified reaction time. After reaction, the support was taken out, washed with water and dried at 80 °C.

## 3. Results and discussion

The FAU zeolite layer formed was characterized by XRD and SEM. Table 1 summarized the properties of NaY zeolite membranes prepared with different gels. The aging effect of the gel is also summarized in Table 2. A room temperature aging is favorable to synthesis NaY zeolite membranes. The crystallization of NaY membranes are strongly influenced by hydrothermal temperature and time. A rise in temperature will increase the grown rate of Y zeolite crystals with a certain time. However, the impure of NaP zeolite, a more stable phase, will form and replace the NaY at high temperature and long time procedure.

High membrane performances were observed in pervaporation of water/ethanol separation as shows in Table 3. High performance Y-type membrane was prepared with gel of $SiO_2$ : $Al_2O_3$ : $Na_2O$ : $H_2O$=15 : 1 : 13.5 : 1215. A short treatment time would be favorable to growth of pure membranes.

1) H.Kita, “Zeolite Membranes for Pervaporation and Vapor Permeation” in “Materials Science of Membranes” Ed. by Y.Yampolskii, I.Pinnau B.D.Freeman, Wiley, 2006, p373-389.

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Continuous and dense Y zeolite membranes were synthesized by two steps of heating procedure, such as at first a high temperature (140 °C) and then a lower temperature (100 °C).

Table1. Properties of NaY zeolite membranes prepared with different gels

| Gel No. | Gel composition $SiO_2$: $Al_2O_3$: $Na_2O$: $H_2O$ | Membrane layer | PV results (10 wt% water/ethanol) | |
|---|---|---|---|---|
| | | | Q/kg·m$^{-2}$·h$^{-1}$ | α |
| G1 | 20:1:8:400 | - | no | no |
| G2 | 15:1: 13.5:1215 | Y | 2.51 | 140 |
| G3 | 20:1:12.5:800 | Y | 2.90 | 60 |
| G4 | 12.8:1:17:975 | Y+P | 1.97 | 84 |
| G5 | 10:1:14:800 | Y+P | 1.88 | 110 |
| G6 | 25:1:22:990 | Y | 1.86 | 125 |

Note: [a] Aging time 12 h; Support: mullite tube; [b] Synthesis: 110 °C. 6 h: [c] PV: 75 °C. $H_2O$/EtOH(10 wt%/90 wt%) feed.

Table2. PV performance of NaY zeolite membranes synthesized after different aging time

| Membrane No. | Aging time(h) | Flux(kg·m$^{-2}$·h$^{-1}$) | Separation factor |
|---|---|---|---|
| M09 | 0 | 3.10 | 35 |
| M10 | 6 | 1.75 | 70 |
| M11 | 12 | 2.26 | 96 |
| M12 | 18 | 1.94 | 210 |
| M13 | 36 | 1.82 | 63 |

Note: [a] PV test at 75 °C, $H_2O$/EtOH(10 wt%/90 wt%) feed;
[b] Gel of $15SiO_2$:$1Al_2O_3$:$yNa_2O$:$1215H_2O$;
[c] Synthesized for 6 h at 110 °C on mullite.

Table3. PV performance of NaY membranes prepared from different temperature

| | Synthesis | | PV | |
|---|---|---|---|---|
| | Temp (°C) | Time (h) | Flux kg/m$^2$h | α |
| M34 | 100 | 6 | 2.5 | 130 |
| M35 | 110 | 6 | 2.23 | 245 |
| M36* | 110 | 6 | 5.06 | 220 |
| M37 | 140, 100 | 1.5, 3 | 2.19 | 300 |
| M38* | 140, 100 | 1.5, 3 | 4.70 | 270 |
| M39 | 100, 140 | 3, 1.5 | 1.50 | 75 |

Note: *on SS tube; [a] Gel, $15SiO_2$:$Al_2O_3$:$13.5Na_2O$:$1215H_2O$, aged 18 h; [b] PV tests for 10 wt% $H_2O$ /90 wt% EtOH at 75 °C.