論　文　の　要　旨

題　目　　Development of fluorine induced microporous silica membranes for carbon dioxide separation

（CO_2分離のためのフッ素含有シリカ系多孔膜の開発）

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Membrane gas separation offers appealing characteristics of low energy consumption, compact size, simply in process, and cost effectiveness over the other conventional separation methods such as cryogenic separation and amine absorption. More specifically, silica membranes outperformed polymeric and zeolites in terms of chemical and thermal stability, high molecular sieving effect, easy synthesis, and cost effectiveness. Hence, the objective of this dissertation was to tailore the network pore sizes of silica membranes for CO_2/N_2 separation under dry/wet conditions.

Firstly, a strategy was proposed to control the network pore structure of conventional silica via fluorine doping to fabricate highly permeable membranes for CO_2 separation. A fluorine-doped silica membrane showed the highest permeance ratio of 50 with CO_2 permeance of 1.6×10^{-7} mol m^{-2} s^{-1} Pa^{-1} at 50 ºC. These results were ascribed to the enlarged pore size as well as the controlled CO_2 adsorption properties. Secondly, a series of pendant alkoxysilane structures with various carbon numbers (C_1-C_8) were used to fabricate sol-gel derived organosilica membranes to evaluate the effects of the C/Si ratio and fluorine doping. Fluorine-doped pendant-type organosilica membranes showed pore sizes like those of undoped organosilica membranes and it was attributed to the flexibility of pendent side chain which hindered the effectiveness of fluorine. Finally, amide-functionalized silica membranes were fabricated for CO_2/N_2 separation via the use of tetraethoxysilane (TEOS) and amine-silica (3-aminopropyltriethoxysilyl, APTES) precursors in the presence of trifluoroacetic acid (TFA). The developed composite membranes exhibited a CO_2 permeance

of 3.8 × 10^{-7} mol m^{-2} s^{-1} Pa^{-1} and a CO_2/N_2 selectivity of 35 at 50 ºC during binary gas separation, which was facilitated by a functionalized dual network structure with size-sieving properties (tailored network) and CO_2-philic sites (amides).