Development of Mechanically Reinforced Free-standing Nanomembranes

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Name

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（強度増強された自立ナノ膜の開発）
Title

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論　文　内　容　の　要　旨

Attempts to develop highly compliant free-standing nanomembranes (FS-NMs) with unprecedented macroscopic mechanical stabilities remained a challenging task. In this thesis entitled as above, a research work aiming to address the toughing of FS-NMs which is essential for practical applications has been presented. In the course of this research, an effort has been put to develop a facile strategy for the preparation of free-standing and mechanically strong, yet functional sub-100 nm thick membranes. Organic/inorganic composite materials are the subject of this research as they pave the way to synergetic mechanical reinforcement by combining the conflicting features of flexibility in polymers and rigidity in inorganics. The developed nanomembranes were tested for gas separation, which is among the most complex molecular separation processes due to its hypersensitivity to leakage. The achievements during the course of this research work has been outlined as follows.

Firstly, a metal oxide/polymer composite nanomembrane has been prepared by a sol-gel induced LBL assembly and discussed in Chapter 2 of the thesis. The spin-assisted LBL assembly of poly(vinyl alcohol) (PVA) and TiO_2 was aimed to maximize the synergetic mechanical enhancement between the polymer and metal oxide via strong sol-gel reactions. Accordingly, a few tenths nm thick, large size, highly flexible and free-standing composite nanomembrane was developed. The nanomembrane sustained mechanical stability even after transfer onto a porous support for separation experiment. The electron microscopy observation proved no visible cracks in the nanomembrane. However, the membrane demonstrated low preferential separation of gas mixtures. The little or no selective transport could be due to the formation of pinholes either during membrane preparation or processing steps. This, in turn, would mainly be associated to the hard nature of PVA, which doesn't soften the oxide phase sufficiently in the composite nanomembrane. In fact, PVA doesn't have specific affinity to CO_2 which was the subject of separation.

In chapter 3, two measures were investigated to assess the low separation performance (which reflects unsatisfactory mechanical stability) of the PVA/TiO_2 LBL nanomembrane. (i) Choosing a softer polymer with better gas separation properties, and (ii) changing the membrane architecture. Accordingly, a blended type of PEG/TiO_2 FS-NM was developed from

a blend formula of a hydroxyl-terminated polyethylene glycol (PEG-OH) and silicon tetraisocyanate. Although no significant mechanical property enhancement was observed, compared to the PVA/TiO_2 LBL nanomembrane, the PEG/SiO_2 hybrid nanomembrane demonstrated improved CO_2/N_2 selectivity.

The obtained results in Chapter 2 and Chapter 3 implied that it is not smooth to improve mechanical property of a nanomembrane while maintaining the separation performance or vice versa only by changing membrane materials. Of course, there can be a probability to tune both mechanical property and function together. It would, however, be time consuming and costly to search for the best material combinations. Therefore, a more reliable membrane architecture and preparation method was still needed.

Accordingly, an unconventional design approach that integrates macroscopic mechanical stability together with tunable membrane functions was developed and presented in Chapter 4. This unique strategy involves the incorporation of aluminosilicate nanotube (ASNT) network structure as a bed scaffold to deposit polymeric membranes. Unlike amorphous metal oxides, aluminosilicate nanotubular mixed metal oxide scaffolds brought about a dramatic enhancement in the mechanical property of the composite nanomembrane with polydimethylsiloxane (PDMS). A four-fold increase in the tensile strength and over 43 times higher biaxial modulus was observed compared to the pristine PDMS. Expansion of the established method to the preparation of various polymer-based FS-NMs has also been presented.

The dissertation presented a significant step-forward in the realization of ultrathin and free-standing, yet macroscopically stable advanced membranes for practical applications, such as small molecule separations.

〔作成要領〕

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