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Rapid N₂/O₂ separation via graphene-zeolite interfacial 2D nanochannels

Energy-friendly separation methods, such as membrane and adsorption separation, have been requested to construct a sustainable society. Recently, we have developed an ultra-high-speed gas separation membrane, called a graphene-wrapped MFI zeolite (Gr/MFI) membrane^[1]. The Gr/MFI membrane separates gas molecules via interfacial nanochannels between zeolite crystal and graphene sheet and exhibits superior gas separation properties. Control of the interfacial 2D nanochannels is the key to achieve high separation performance. The Gr/MFI membrane is obtained through wrapping of zeolite crystals with graphene oxide (GO) in the colloidal state. The electrostatic interactions between GO and zeolite crystals dominates the 2D interfacial nanochannel. Control of surface property of unstable GO colloids^[2] is also an important factor.

In this presentation, we demonstrate superiority of intrinsic GO as a nanocrystal-wrapping agent. Stable and efficient wrapping of zeolite nanocrystals with intrinsic GO provides interfacial nanochannels suitable for N₂/O₂ separations. The Gr/MFI membrane obtained from intrinsic GO exhibits an ideal separation factor for N₂/O₂ of 2.6 with the N₂ permeability of the order of 10⁵ Barrer, which is three orders faster than the polymer-based and inorganic membranes. The N₂/O₂ selectivity of Gr/MFI membrane is improved by the control of the electrostatic interactions^[3], associated with the structure of GO-wrapped MFI zeolites.

[1] R. Kukobat, M. Sakai, H. Tanaka, H. Otsuka, F. Vallejos, C. Lastoskie, M. Matsukata, Y. Sakai, K. Yoshida, T. Hayashi, K. Kaneko, *Sci. Adv.* 8, eabl3521, 1-11 (2022).

[2] H. Otsuka, K. Urita, N. Honma, T. Kimuro, Y. Amako, R. Kukobat, T. J. Bandosz, J. Ukai, I. Moriguchi, K. Kaneko, *Nat. Commun.* 15, 1708, (2024).

[3] H. Otsuka, K. Kubo, E. Z. P. Salazara, A. Furuse, Y. Yoshikawa, R. Kobayashi, K. Kaneko *Next Mater.* **in Revision**.