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Unveiling a differential-sorption mechanism for enhanced CO_2 capture: Highly selective Fe_2O_3- and Fe_3O_4-based polyimide mixed-matrix membranes

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This study presents a comprehensive investigation of polyimide (PMDA-ODA) nanocomposite membranes incorporating hematite (Fe2O3) and magnetite (Fe3O4) nanoparticles. Incorporation of these fillers primarily aims to reduce nitrogen permeability by reducing the adsorption of this gas on the membrane surface, thereby enhancing membrane selectivity for the CO2/N2 system. The gas separation performance was evaluated using pure CO2 and N2 permeation tests, which systematically determined key transport parameters, including permeability, diffusion coefficients, solubility coefficients, and ideal selectivity. The optimal filler content was identified as 5 wt% for Fe2O3 and 2 wt% for Fe3O4, achieving the highest ideal selectivity values of 76.45 and 98.08, respectively. At these loadings, the corresponding CO2 permeabilities were 2.60 and 3.07 Barrer. The principal novelty of this work is the introduction and comprehensive investigation of a gas-transport mechanism referred as differential-sorption, which has been applied for mixed matrix membranes for the first time. The transport model was developed to better understand the transport behaviour depending on filler type and loading in polymer matrix. A fundamental assumption of this mechanism is limited sorption of one studied gases, while the second gas transport is stabilized by improved interactions with filler and Knudsen diffusion in magnetic channels. The model accuracy was confirmed through random-walk simulations and benchmarked against experimental results and classical predictions from the Maxwell and Bruggeman models. The strong agreement between

the model and experimental data was further supported by detailed structural and property analyses, including thermogravimetric analysis, scanning electron microscopy, atomic force microscopy, gas sorption studies, and mechanical and magnetic measurements.

Metryczka

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