

Nanoporous Graphene Membrane for Health and Environmental Applications

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Separation processes are found in many diverse applications, including health (drug purification, sterilization) and environment (CO₂ capture, water treatment, resource recovery). Compared to thermal-based separation systems, membranes are modular and have the potential for more efficient separations without the need for extreme temperatures and large equipment. However, conventional polymeric membranes can suffer from fouling, poor stability at high temperatures and in harsh chemical environments, are subject to a permeability/selectivity trade-off, and it remains hard to precisely control and engineer their structures on the molecular level. These limitations call for the development of new membrane materials to yield significant performance improvements.

The emergence of 2D nanomaterials allows for the creation of atomically thin membranes such as nanoporous graphene (NPG), and offers the opportunity to enhance chemical stability as well as increase both permeability and selectivity via significantly reducing membrane thickness and controlling the pore structure. Despite significant progress in theoretical and experimental work in the development of NPG membranes, challenges remain to be addressed for NPGs to be deployed, particularly in the control of leakages through defects, the limited experimentally-supported transport understanding, and the exploration and design of NPG systems for various health and environment applications.

This thesis extends the theoretical understanding of transport across graphene composite membranes and demonstrates how differences in the scaling of transport rates with pore size for viscous flow, gas effusion, dilute solute diffusion, and ion transport, together with the interplay between graphene and support structure and selective pore size distribution, influence leakage and selectivity. This thesis also explores how non-linear current-voltage relationships can arise in ultra-thin membranes due to induced charge effects. The models developed using the theories enable us to estimate permeation rates without the need for computationally-intensive simulations.

Furthermore, this thesis presents the application of NPG to hemodialysis, desalination, and ion separations. For dialysis, using system level modeling of the device and its interaction with the body, the thesis shows that current dialyzers are membrane mass-transfer limited for protein-bound uremic toxins (PBUTs), and establish performance targets for NPG membranes to enhance PBUT removal. These targets are translated to the novel design and fabrication of an NPG composite membrane. The thesis also investigates the desalination performance achievable by NPGs with pore size distributions that are practically realizable and ways to surpass the polymeric permeability/selectivity trade-off limit, and develops a novel experimental/analysis procedure to study simultaneous ion transport across NPG and strategies to enhance selectivity for the recovery of rare earth elements.

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