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Graphene-induced Tuning of the d-Spacing of Graphene Oxide Composite Nanofiltration Membranes for Frictionless Capillary Action-induced Enhancement of Water Permeability

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2.5. Computational investigation.

The mean square displacement (MSD) of a molecule refers to the average distance it travels. According to diffusion theory, MSDs are proportional to the elapsed time with power α, t^{α}. Different α indicate different types of diffusion. When α is equal to one, the diffusion character is called normal diffusion. Self-diffusion coefficients are obtained by calculating the slope of MSD_S. When molecules travel in three different dimensions, the self-diffusion coefficients are equal to the slope of the MSDs divided by 6. In the system under study, water molecules only travelled in a plane, therefore the self-diffusion coefficients were equal to the slope of the in-plane MSDs (excluding the surface direction) divided by 4. When α is greater than 1, molecules undergo superdiffusion, which means the molecules spread faster than in normal diffusion. In-plane MSD calculations were performed by the NVT ensemble, and volume simulation boxes were obtained by 4 ns simulations through an NPT ensemble. The Nose–Hoover chain barostat and thermostat were implemented for the NPT and NVT ensembles. The relaxation times of the barostat and thermostat were set to 1 and 0.1 ps, respectively, the temperature and pressure were set to standard conditions (298.15 K and 1 atm, respectively), and the equation of motion was integrated by the Velocity–Verlet algorithm with a time step of 1 fs. Coulomb interactions were evaluated by the particle–particle–particle–mesh method, and the cutoff for directly evaluating Coulomb interactions was 12 Å. Twelve independent simulations were conducted to collect in-plane MSD data for each GO–graphene layer model, and each simulation lasted for 35 ns. These 12 in-plane MSD data were averaged for further evaluation as shown in **Fig. S1**. The left model contains four GO layers. The middle and right models are mixed GO–graphene layers and contain two graphene layers and two GO layers. The graphene and GO layers are arranged alternately in the GO,graphene,GO,graphene model, and the graphene and GO layers are placed

separately in the GO,GO,graphene,graphene model. The space between layers is filled with water molecules, which are represented, and the layer spacing is 13–14 Å. Black horizontal lines indicate the graphene layers, and blue Os indicate the epoxy groups on the GO surfaces.

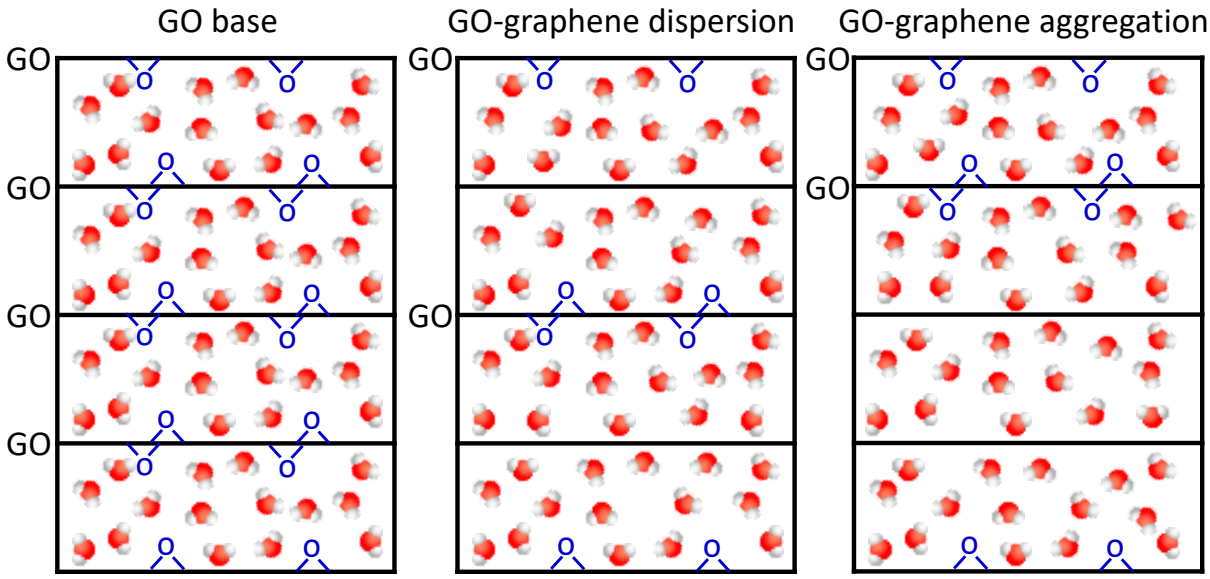

Fig. S1. Schematic of three different mixed GO–graphene layer models used in molecular dynamics simulations.