Supporting Information

Graphene foam membranes with tunable pore size for next-generation reverse osmosis water desalination

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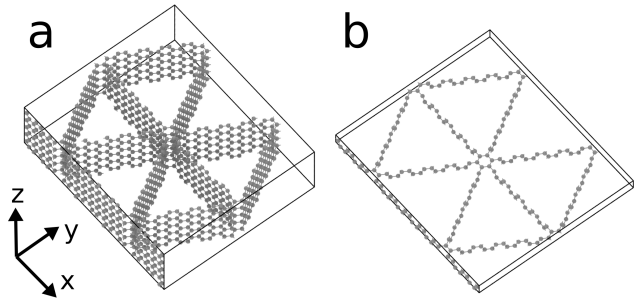

Figure S1: (a) Molecular dynamics and (b) first-principles models of a graphene foam membrane with an initial pore area of 187 Å^2.

We calculate Young's modulus and Poisson's ratio of a graphene foam membrane with an initial pore area of 187 Å^2 using a molecular dynamics simulation and a first-principles calculation based on density functional theory. The sizes of the orthorombic simulation boxes are 41.8 Å × 36.2 Å × 12 Å (Figure S1a) and 41.8 Å × 36.2 Å × 2.5 Å (Figure S1b). Periodic boundary conditions are applied in all directions. In the molecular dynamics simulation the system initially is relaxed at 1 K for 100 ps using an isothermal-isobaric ensemble. Afterwards compressive strain is applied in the x-direction with a rate of 10^8 s^{-1} and the resulting stresses in the y- and z-directions are controlled to be zero using an isothermal-isobaric ensemble (to simulate uniaxial stress in the x-direction). In the first-principles calculation an energy cutoff of 600 eV is applied and the reciprocal space is sampled on a 1 × 1 × 20 k-mesh based on the Monkhorst-Pack scheme [1]. The system initially is relaxed with a force convergence

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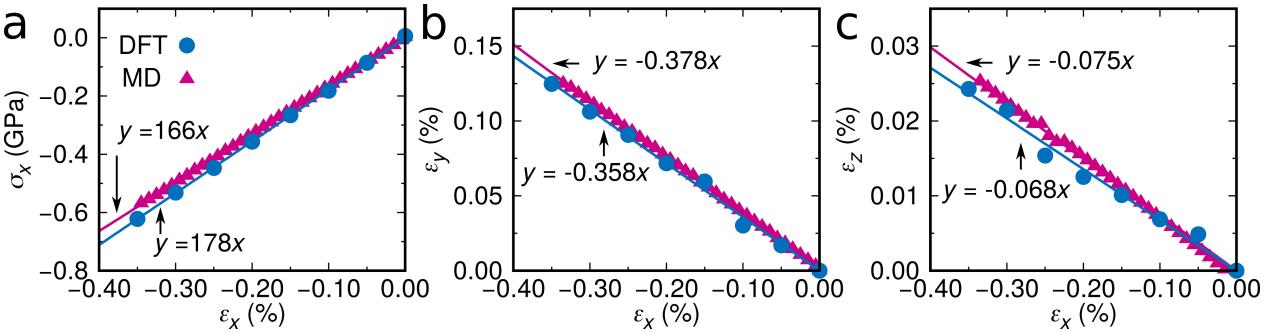

Figure S2: Comparison between the molecular dynamics simulation and first-principles calculation of a graphene foam membrane with an initial pore area of 187 Å^2.

criterion of 0.01 eV/Å. Thereafter compressive strain is applied in the x-direction in steps of 0.0005 and the system is relaxed along the y- and z-directions (to simulate uniaxial stress in the x-direction) using the same force criterion (Vienna Ab initio Simulation Package [2] with projector augmented wave method [3], Perdew-Burke-Ernzerhof exchange-correlation functional [4], and van der Waals correction [5]). Eventually, Young's modulus is obtained as the slope of the linear fitting curve of the stress as a function of the applied strain (Figure S2a) and Poisson's ratio is obtained as the slope of the linear fitting curve of the lateral strain as a function of the applied strain (Figures S2b and S2c).

The interlayer binding energy of graphite is obtained by molecular statics simulation and first-principles calculation as $\gamma = (e - e_g)/A_g$, where e is the energy per atom of graphite, e_g is the energy per atom of graphene, and A_g is the area per atom of graphene. In the molecular statics simulation we assign the x-, y- and z-directions to be the zigzag, armchair, and out-of-plane directions, respectively. The sizes of the simulation boxes are 24.2 Å × 20.9 Å for graphene and 24.2 Å × 20.9 Å × 20.2 Å for graphite. Periodic boundary conditions are applied in the in-plane directions for graphene and in all directions for graphite. The systems are relaxed using the conjugate gradient method untile the relative total energy change between successive iterations is less than 10^{-16}. In the first-principles calculation the hexagonal primitive unit of graphene with an in-plane lattice parameter of 2.46 Å is augmented by a 20 Å thick vacuum layer in the z-direction. The same in-plane lattice parameter is used for graphite with an out-of-plane lattice parameter of 6.70 Å. 16 × 16 × 1 and 16 × 16 × 8 k-meshes based on the Monkhorst-Pack scheme are chosen for graphene and

graphite, respectively. An energy cutoff of 600 eV is used with a force convergence criterion of 0.01 eV/Å. Periodic boundary conditions are applied in all directions (Vienna Ab initio Simulation Package [2] with projector augmented wave method [3], Perdew-Burke-Ernzerhof exchange-correlation functional [4], and van der Waals correction [5]).

Our results show that the mechanical properties of the graphene foam membrane obtained by the molecular dynamics simulation using the AIREBO potential agree well with those obtained by the first-principles calculation (Figure S2). The Young's modulus in the x-direction and Poisson's ratios ν_{xy} and ν_{xz} turn out to be 166 GPa, 0.378, and 0.075 in the molecular dynamics simulation and 178 GPa, 0.358, and 0.068 in the first-principles calculation. Furthermore, the interlayer binding energy of graphite (being essential for the graphene skeleton) gives the same value of 32 J m^{-2} in the molecular statics simulation and first-principles calculation, being close to the experimental value of 33 J m^{-2} [6].

References

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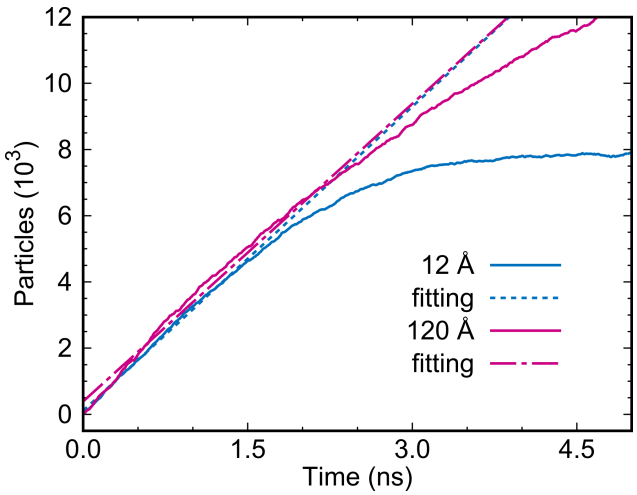

Figure S3: Number of water molecules passing through unstrained honeycomb graphene foam membranes with a common initial pore area of 69 Å^2 and thicknesses of 12 Å and 120 Å as a function of time. The membranes provide perfect salt rejection with water permeabilities of 152 and 150 l cm^{-2} day^{-1} MPa^{-1}, respectively.

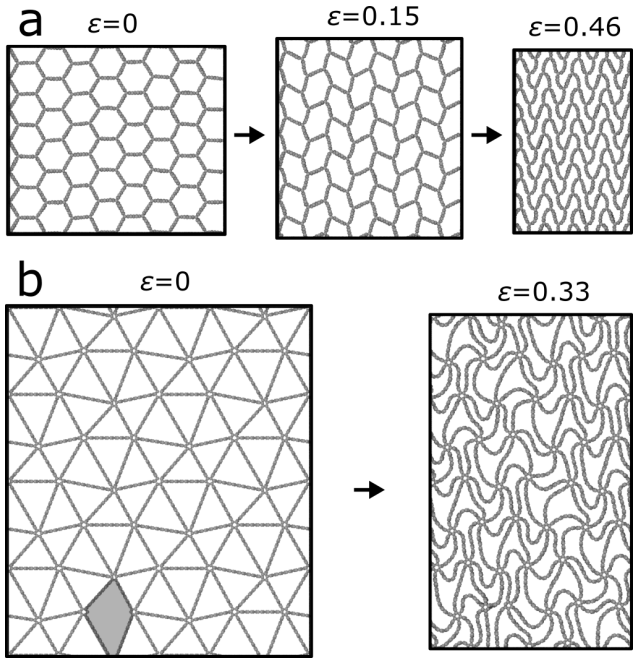

Figure S4: (a) Honeycomb graphene foam membrane with an initial pore area of 150 Å^2 and a thickness of 12 Å at different strains applied in the x-direction. (b) Non-uniform graphene foam membrane with different initial pore sizes and shapes and a thickness of 12 Å at different strains applied in the x-direction. The initial area of the triangular pores ranges from 168 to 231 Å^2 and that of the gray shaded pore is 399 Å^2.