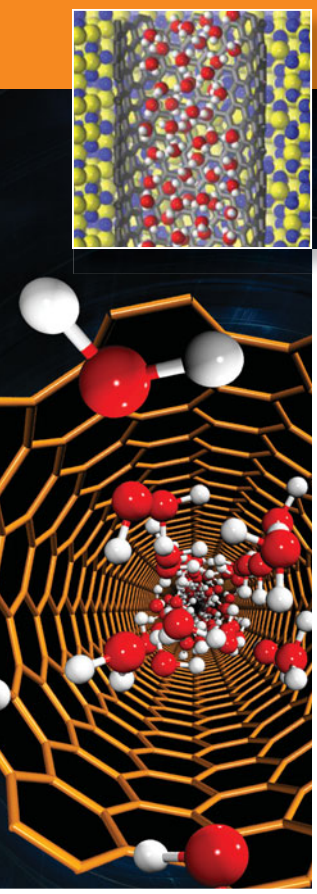


Multiscale simulation of enhanced water flow in nanotubes

Matthew K. Borg and Jason M. Reese

Nanotubes (NTs) with diameters less than 2 nm have been proposed for next-generation reverse osmosis membranes. At this molecular scale, the NTs are narrow enough to block salt ions and other contaminants, but still wide enough to allow water to flow along the NTs at seemingly unprecedented rates. Simulations for design of NT membranes can be challenging. On the one hand, the standard equations for water flow through pipes are not applicable at sub-2-nm scales due to the dominance of non-continuum phenomena; on the other hand, full molecular simulations are computationally intractable for flows up to laboratory or prototype scales. This article describes recent multiscale approaches to simulating flows through aligned NT membranes of various materials. These multiscale techniques offer a unique and economical solution that can shed light on sometimes conflicting experimental results and point the way to future engineering design of nanostructured membranes.

Introduction

Drinking water scarcity is a major and increasing problem in both developing and developed nations. However, seawater and brackish water are abundant. Reverse osmosis (RO) desalination—the process of forcing seawater through a semipermeable membrane to remove salt ions and other contaminants—is one of the most promising routes toward large-scale freshwater reclamation.

Current commercial thin-film composite RO membranes are made from polymeric materials (typically polyamide), but there is great scope to develop advanced nanostructured RO membranes to mitigate energy and capital costs. These include nanoporous membranes built on ceramic–inorganic or metal–organic frameworks (such as zeolites), graphene membranes with precisely controlled porosity, biological membranes (e.g., aquaporin proteins), and membranes comprising aligned nanotubes (NTs).[1]

In this article, we focus on water flow through NTs, because of their high reported pure water permeabilities and controllable selectivity. The high flow rates arise from the smooth (low friction) inner surfaces of the NT material. For example, the internal surface/water friction of a carbon nanotube (CNT) has been found to decrease with decreasing diameter: the friction is graphene-like for CNT diameters $D \gtrsim 20$ nm, but falls to almost zero for $D = 0.8$ nm.[2] The selectivity of ions and other dissolved material is also sensitive to the NT diameter, with near 100% salt ion rejection occurring for CNTs with a diameter of ~0.8 nm, decreasing rapidly to 0% when the diameter increases to ~2 nm.[3]

Research into flows through NTs of different materials—such as carbon, boron nitride, and silicon carbide—is now just more than a decade old, but commercial membranes of aligned NTs are still not available. This is mainly due to the difficulty in manufacturing these membranes inexpensively in as close to their ideal form as possible—aligned, pristine NTs encapsulated in a robust, defect-free matrix, with a strictly controlled pore entrance chemistry and pore-size distribution, and a large active pore density.

Flows through NT membranes: A multiscale modeling challenge

Computational modeling of flows through NT membranes is a challenging problem; however, when combined with experiments, it can play a vital role in this materials research area. Simulations can provide useful scientific insights, for instance, into the large disparity in experimental results. They can also be used to quickly explore a much larger parametric space than would be feasible using experiments (e.g., various materials, operating pressures, NT diameters, and membrane thicknesses). Additionally, membrane simulations enable new ideas and concepts to be tested before launching complex experimental campaigns (e.g., modifying the NT inlet/outlet pore chemistry to control performance).

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The major challenge in modeling flows through NT membranes is that conventional fluid models break down at nanometer-length scales ($D \lesssim 2$ nm). For example, using the no-slip Hagen–Poiseuille (HP) fluid equation to calculate the flow rate through a NT has been found to underpredict experimental results for NTs by two to five orders of magnitude;[4–7] there is a substantial NT flow increase above what is expected. The "flow enhancement factor," E, is often used to compare the observed flow rate, $\dot{m}$, with conventional predictions:

$$E = \dot{m}/\dot{m}_{\mathrm{HP}},$$

where the no-slip HP flow rate is:

$$\dot{m}_{\mathrm{HP}} = \frac{\pi D^4 \rho \, \Delta P}{128 \, \mu \, L}. \quad (1)$$

Here, D is the NT diameter, ρ is the fluid density, ΔP is the overall applied pressure drop, μ is the fluid viscosity and L is the length of the NT. The large flow enhancement observed in NTs is thought to be due to the different molecular and non-continuum flow behavior when water is highly confined within sub-2-nm diameter tubes, including slip at the tube walls, molecular ordering, and nonlocal viscosity—all of which may also be diameter dependent. Even with adjustments for fluid slip (e.g., the Navier slip equation), the HP model of Equation 1 still does not operate well for $D \lesssim 2$ nm, as it fails to capture these crucial non-continuum phenomena.[8]

Molecular dynamics (MD) simulations, in which the movements and interactions of all the NT and fluid molecules are tracked in time and space, produce higher fidelity results. However, the NTs used in membrane experiments are generally μm–mm long, with active areas a few μm².[5] These dimensions are too large to be treated by simulations alone, even using the fastest supercomputers.

Multiscale flow modeling is an economical and accurate alternative that exploits smaller and computationally less expensive molecular simulations to solve larger problems in time and space.[9–11] **Figure 1** shows an example of how an ideal NT membrane of laboratory dimensions can be treated using a multiscale technique. The key is to identify features of the membrane that are scale-separated in both time and space and do not require molecular simulation. This enables the molecular resolution to be reduced; fewer molecules and time steps are needed in the simulation overall, so large computational savings can be achieved.

An individual NT is a high-aspect-ratio conduit. Non-continuum fluid effects mainly occur across the cross section of the tube, and these are scale-separated from variations in the flow direction. Macroscopic streamwise pressure and velocity change gradually along the NT and are much slower processes than the microscopic agitation of molecules that are costly to resolve by MD. The flow through the NT can, therefore, be modeled without much loss in accuracy, by representing the NT itself as smaller MD sections of length ΔL (as shown in Figure 1), that are coupled with one-dimensional (1D) equations of fluid mass continuity and momentum conservation applied in the streamwise direction alone.

This multiscale treatment is called the internal-flow multiscale method (IMM),[11–13] and has been created in order to deal with high-aspect-ratio flows that are characteristic of liquids in NTs, rarefied microgas applications (such as MEMS pumps,[9,10] lubricating bearings,[13] and microcrack flows[14,15]) and other systems. In the IMM, the coupling between microscopic MD and macroscopic (1D fluid) models occurs during the simulation. The macroscopic

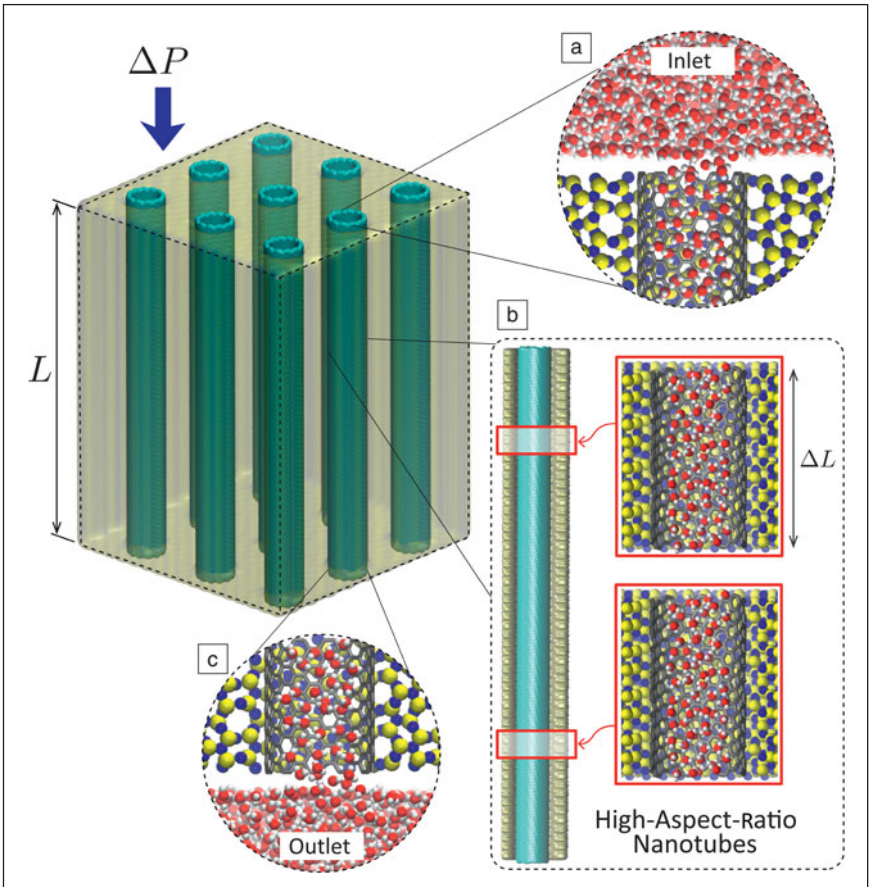


Figure 1. Schematic of multiscale model of a filtration membrane comprising aligned carbon nanotubes within a silicon nitride matrix, similar to the experimental setup in Reference 5. The major contributors to pressure losses (ΔP) in this ideal system are (a) membrane inlets, (b) friction in high-aspect-ratio nanotubes, and (c) membrane outlets. L is the length of a nanotube and ΔL is a representative streamwise length of the molecular dynamics subdomain of the nanotube.

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model assigns the local pressure gradients to each MD subdomain via microscopic forces. These then produce individual flow rates in the MD subdomains that correct the governing continuity and momentum equations, which, in turn, supply new forces back to the MD subdomains. This process repeats until, in steady state, the flow through all MD subdomains is exactly the same. The result is the solution to the problem.

The key advantage of a multiscale formulation such as this is that it is free of any of the constitutive or boundary approximations[16] that make other computational methods invalid. The downside is that the spacing between the MD subdomains needs to be found by trial and error—although this would correspond to the grid- and time-dependency studies that are required to ensure accurate conventional computational fluid dynamics (CFD) solutions. For water flowing along a NT, the strong hydrogen bonds in water ensure incompressibility, even when molecules pass single-file along the tube (i.e., when D = 0.81 nm); so usually only one MD subdomain is needed to accurately describe any length of NT conveying water. This results in enormous computational savings of $\sim L/\Delta L$, where $L \sim$ μm–mm is the length of the NT (or the thickness of the aligned NT membrane) and $\Delta L \sim$ nm is the representative streamwise length of the MD subdomain in the NT. The computational savings over a full MD simulation of the entire NT can be as high as a factor of $\sim 10^6$.[10,11]

Thinner NT membranes (i.e., shorter NTs) experience relatively large pressure losses at the inlet and the outlet of the membrane.[17] In these multiscale simulations, the inlet/outlet regions (see Figure 1a,c) need to be resolved using MD as well, and connected seamlessly within the IMM, for better prediction of the overall mass flow rate.[18,19] The inlet's inclusion in the multiscale model is in any case needed in order to capture the molecular detail of the steric hindrance that is the basis of the membrane's filtration capabilities.

The simulation results we present here assume steady water flow in ideal membranes with perfectly aligned and distributed NTs of equal diameter; our results are for just one NT taken from a section of the membrane, which is sufficient to make flow predictions for the whole membrane. The IMM technique can also be applied to nonideal scenarios, including gradual variations in the cross section of the nanoscale conduit,[12] unsteady/time-dependent flows,[10,11] conduits with local wall defects,[18] general networks of connected and bifurcating channels,[20] as well as vacancy defects distributed along the NT.[21]

Flow results for NTs of different materials

In a multiscale flow simulation, the necessary coupling between the MD simulation and the continuum fluid formulation can be of three forms—concurrent (i.e., the MD simulations run continually with the continuum simulation),[11,19] sequential (i.e., MD presimulations first generate flow data as multidimensional libraries/interpolants, which are then used by subsequent continuum simulations),[8,22] or adaptive sequential/concurrent (i.e., using machine learning to switch optimally between the concurrent/sequential approaches).[23]

We now present multiscale results from both concurrent and sequential simulations of pressure-driven water flows through NTs of different materials, but with fixed pressure drops and similar diameters:[19,24] CNTs with D = 2.034 nm, boron nitride nanotubes (BNNTs) with D = 2.072 nm, and silicon carbide nanotubes (SiCNTs) with D = 2.062 nm (see **Figure 2**a). In the absence of reliable experimental results, full MD simulations are used as benchmark solutions for NTs

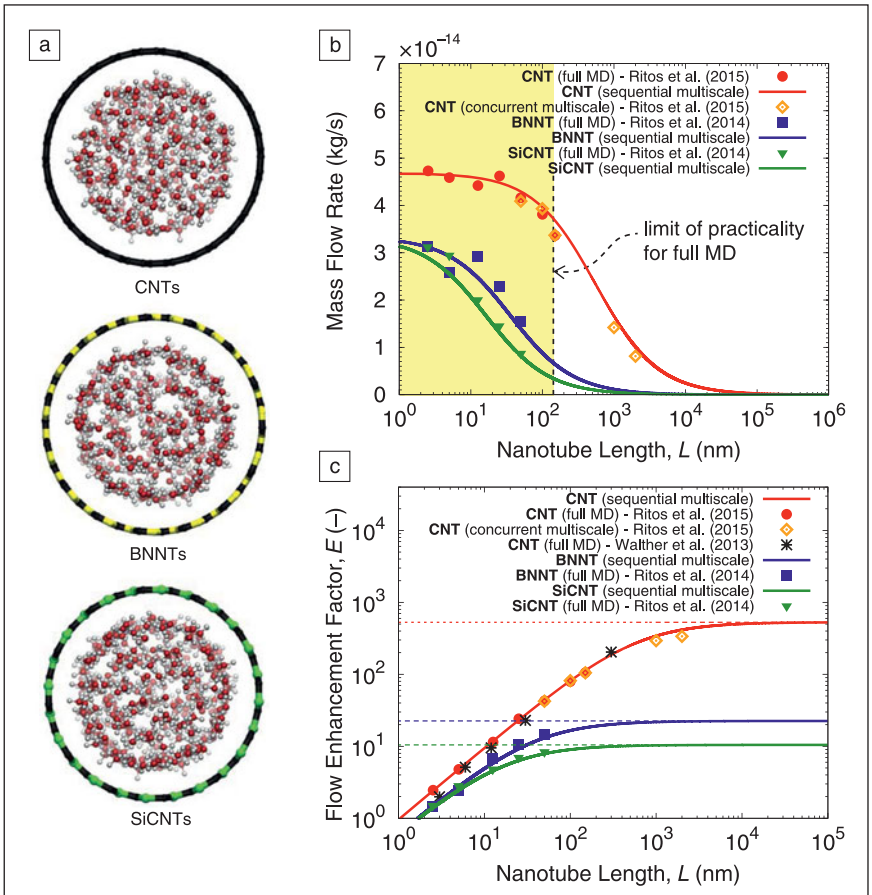


Figure 2. Molecular dynamics (MD) and multiscale simulations of pressure-driven water flow in nanotubes (NTs) of three different materials. (a) Cross sections of the water molecules in carbon nanotubes (CNTs), boron nitride nanotubes (BNNTs), and silicon carbide nanotubes (SiCNTs).[24] Simulation results for (b) mass flow rate and (c) flow enhancement with varying NT length (i.e., membrane thickness). Full MD results are presented up to L = 150 nm. Multiscale methods enable thicker membrane sections to be simulated, and are indicated here by solid lines (for sequential coupling results) and open symbols (for concurrent coupling results).[19,22,24]

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up to around $L = 150$ nm, beyond which MD is too computationally expensive to be practical.

Figure 2b shows mass flow rate results for a single NT, with increasing NT length L up to around 1 mm. There is generally good agreement between the MD comparison simulations (filled symbols) and the multiscale simulations (lines and open symbols) for the three NT materials. As these three cases are for NTs of nearly the same diameter, Figure 2b also indicates clearly that the NT material plays an important role in the flow behavior. CNTs seem to exhibit the lowest internal surface/water friction, which is evident from the higher flow rates and the persistent tail in the flow rate curve as the membrane gets thicker. BNNTs and SiCNTs, however, have higher surface/water friction. This is possibly due to the "rougher" interaction energy landscape experienced by water in contact with a mixed chemical species NT, as opposed to the CNT, which is a single chemical species.[2] This behavior has also been demonstrated in the variation of the flow enhancement factor with NT length (i.e., membrane thickness), shown in Figure 2c. In all three materials, the losses within the NT are minimal for the thinner membranes ($L \lesssim 20$ nm), and the dominant losses occur at the inlet—so the enhancement factor increases steadily with tube length. For the thicker membranes typical of experiments and lab prototypes, the small but finite friction in the tubes generates most of the pressure losses, so the main pressure loss shifts gradually from the inlet of the NT to the central part of the tube as the NT lengthens. This explains the leveling off of the flow enhancement factor at an upper limit (the horizontal dotted lines in Figure 2c).[19,22] This upper limit is the same as for a flow in an infinitely long NT (i.e., with no inlet/outlet considered) and is the largest possible enhancement that can be achieved for a given D.

The benefit of these multiscale simulations over full MD now becomes clear. Full MD simulations of NTs of laboratory lengths are computationally intractable, while smaller periodic MD simulations representing "infinitely long" NTs only provide the maximum E (they do not provide information about the variation of E with L, or of $\dot{m}$ with L). Only by using a multiscale simulation is it possible to provide insight into the performance of a particular membrane configuration (i.e., with a given NT material of diameter D and length L, and with known inlet/outlet topography and chemistry), and to determine whether it is operating at its maximum flow enhancement or not. Our maximum predicted enhancement factors are: $E = 530$ for CNTs when $L \gtrsim 10$ μm, $E = 22$ for BNNTs when $L \gtrsim 2$ μm, and $E = 10$ for SiCNTs when $L \gtrsim 0.7$ μm.

Comparison with experiment

Experiments to date have been carried out predominantly on pressure-driven flows through CNT membranes, mostly because CNTs offer the largest flow enhancement of all synthesized NT materials, while resisting biofouling and bacterial adhesion at the same time.[25,26] The range of measured flow enhancement factors is staggering; there are two to five orders of magnitude in E between CNTs of similar diameters, with no particular evidence of correlation between these experiments. This disparity in the experimental data has been a source of considerable debate over the past decade, with many open questions about the quality of—and possible errors in—the experiments. Reported flow enhancements predicted by MD are less scattered, but differences can be caused by the choice of intermolecular force fields[27] and temperature-control parameters.[28] Multiscale simulations rely on MD to provide the molecular detail and can suffer the same drawbacks.

Figure 3 shows multiscale simulation results of the flow enhancement varying with CNT length for three CNT diameters: $D = 1.36$ nm, $D = 1.59$ nm, and $D = 2.034$ nm. These diameters were chosen because of their prospective importance in water filtration. Qin et al.[7] and Holt et al.[5] are the only reported experiments that seem to exist within this diameter range. Multiscale simulations now enable us to make direct comparisons with these experiments, whereas full MD simulations would be far too time-consuming. Qin et al. conducted an experiment with a 1-mm-long NT. Comparing these results with one data point from MD would take around three centuries on the fastest supercomputer. Our concurrent multiscale simulation, however, required only 16 weeks[19]—a factor of 1000 saving in computation time. The multiscale simulations for Figure 3 used MD subdomains with water/carbon potentials calibrated from independent experiments of droplets on graphitic surfaces.[29] These multiscale results agree reasonably well with the experiment of Holt et al.[5] However, the results of Qin et al. (e.g., see $D = 1.59$ nm) do not agree with the multiscale simulations (or the results of Holt et al. either) by at least two orders of magnitude; there is no understanding currently of why there is such a large discrepancy in these data.

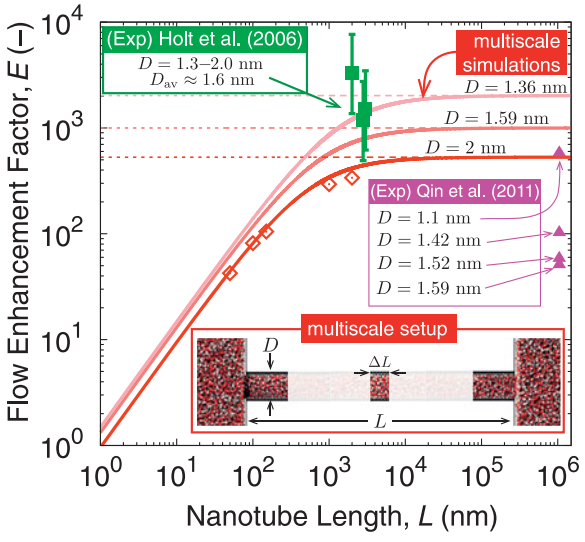


Figure 3. Comparison of sequential (solid lines) and concurrent (open symbols, $D = 2$ nm) multiscale results for the flow enhancement factor with the experimental data of Holt et al.[5] (squares) and Qin et al.[7] (triangles) for carbon nanotubes with diameters as noted. Inset: schematic of the multiscale setup.

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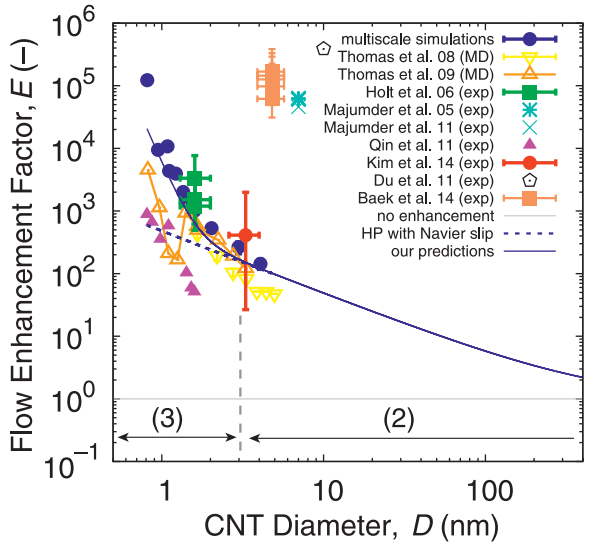


Figure 4. Comparisons of maximum flow enhancement with carbon nanotube (CNT) diameter between various molecular dynamics (MD),[32,33] multiscale,[19] and experimental results.[4–7,25,30,31] Flow regimes (2) and (3) inside the nanotube are also indicated. Note: HP, Hagen–Poiseuille.

Figure 4[4–7,19,25,30–33] shows the influence of CNT diameter on the flow enhancement for thick membranes (i.e., $L \to \infty$; maximum E for any D), including data from molecular dynamics, experiments, and sequential multiscale simulations. The clear trend in Figure 4 is that the flow enhancement E increases with decreasing CNT diameter D, with three regimes observed: (1) no flow enhancement ($E = 1$) for $D \gtrsim 1$ μm (not shown in Figure 4), and the no-slip HP flow model is applicable; (2) flow enhancement varies proportionally to $\sim 1/D$, for 3 nm $\lesssim D \lesssim 1$ μm, and the HP model with Navier slip at the walls is applicable provided the slip length is known (~60 nm for graphene and for CNTs with large D); and (3) flow enhancement increases rapidly for $D \lesssim 3$ nm, and the Navier slip condition with a fixed slip length is no longer valid.

This last regime is pivotal to the success of next-generation RO membranes, but requires knowledge of the unconventional behavior of the slip length as well as the inlet/outlet losses; only multiscale simulations and experiments can provide this insight up to laboratory and prototype scales. The solid blue line in Figure 4 represents results from the established models for regimes (1) and (2), with results from our multiscale simulations in regime (3). While there is reasonable agreement between our multiscale simulations and the experimental results of Kim et al.[30] and Holt et al.,[5] and qualitative agreement in trends with Qin et al.,[7] it is still unclear why there exists such a substantial difference between these and the subset of experimental results[4,6,25,31] clustered in the top part of Figure 4.

Summary

The future of materials for filtration technologies looks bright. Lower costs could be achieved by developing membranes with a high density of aligned NTs and high water flow rates,[34] with evidence pointing toward carbon as the front-runner material. In this article, we have described how advances in multiscale flow modeling mean we can now start to understand the flow of water in NT membranes of different materials, pore diameters, and thicknesses. Our multiscale simulations indicate that the flow enhancement through the NTs (a) increases with NT length until it levels off at a limit (for a given diameter) dependent on the NT material, and (b) increases with decreasing diameter of long carbon NTs in three identifiable regimes.

There is still considerable uncertainty in current experimental and molecular simulation results. In molecular simulations, questions remain about the correct intermolecular potential models to use in high confinement, and the actual slip length of water inside NTs. The wide variance in empirical results means that experiments should be repeated many times in the future, across a wider range of NT diameters, with protocols in place to ensure the NTs are pristine and that the flow rate being measured is that through the actual tubes. What is certain, however, is that multiscale methods have the potential to produce useful results with molecular specificity in both time and space, at a fraction of the computational cost of full MD simulations. Multiscale modeling not only offers a radical and practical opportunity to contribute to the structural and materials design of future membranes, but it can also guide future experimental campaigns for a range of other micro-/nanoflow systems.

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