Membrane Fouling: Microscopic Insights into the Effects of Surface Chemistry and Roughness

Mao Wang, John Wang, and Jianwen Jiang*

Fouling is a major obstacle and challenge in membrane-based separation processes. Caused by the sophisticated interactions between foulant and membrane surface, fouling strongly depends on membrane surface chemistry and roughness. Current studies in the field have been largely focused on polymer membranes. Herein, a molecular dynamics simulation study is reported for fouling on alumina and graphene membrane surfaces during water treatment. For two foulants (sucralose and bisphenol A), it is found that the fouling on alumina surfaces is reduced with increasing surface roughness; however, the fouling on graphene surfaces is enhanced by roughness. It is unravelled that the foulant–surface interaction becomes weaker in the ridge region of a rough alumina surface, thus allowing foulant to leave the surface and reducing fouling. Nevertheless, such behavior is not observed on a rough graphene surface because of strong foulant–graphene interaction. In addition, with increasing roughness, the hydrogen bonds between water and alumina surface are found to increase in number as well as stability. This simulation study reveals that surface chemistry and roughness play a crucial role in membrane fouling, and the microscopic insights are useful for the design of new membranes toward high-performance water treatment.

1. Introduction

In recent years, water scarcity has become a major concern worldwide due to ever-growing population, climate change, and water pollution.[1,2] Clean water is indispensable for global health and economic development. It has become a grand challenge to provide clean water in a reliable and affordable manner. As an energy efficient and cost effective technology, membrane-based water treatment has a wide variety of applications in seawater or brackish water desalination, municipal, or industrial wastewater purification. However, one of the key issues in membrane-based water treatment is membrane fouling. It adversely affects membrane performance by reducing water permeability, deteriorating water quality, lowering membrane lifetime, and increasing energy consumption.[3,4]

Microscopically, fouling is caused by sophisticated interactions between membrane surface and foulant, which strongly depends on the chemistry and morphology of membrane surface. In this respect, surface modification is widely adopted to improve membrane antifouling capacity.[5–7] At present, most studies in this field have primarily investigated the effect of surface chemistry on fouling.[8] Nevertheless, surface morphology or roughness also has a significant impact. A number of experimental and theoretical studies have shown that surface roughness can enhance fouling. For example, Elimelech et al. experimentally found that colloidal particles were preferentially accumulated in the valleys of rough aromatic polyamide thin-film composite membranes, which resulted in "valley clogging" and severely reduced water flux.[9,10] Based on a mathematical model, they explained that the valleys created by surface roughness produced a deeper interaction well in which colloidal particles might preferentially deposit.[11] Using both experimental and simulation techniques, Jun et al. investigated the fouling of poly(methyl methacrylate) (PMMA) colloidal particles on polyvinylidene fluoride (PVDF) membrane surface with periodically spaced prism shape. A stagnant flow zone was found to form in the valley of patterned surface with more particles deposited.[12] On three commercial polyamide thin-film composite reverse osmosis (RO) membranes with different roughness and hydrophilicity, Yin et al. experimentally observed that the fouling was enhanced with an increase in surface roughness.[13]

Interestingly, the opposite effect of surface roughness on fouling (i.e., reduced fouling) was also reported. Gohari et al. fabricated flat sheet poly(ether sulfone) (PES) membranes with aligned patterns by incorporating hydrophilic nanoparticles for the ultrafiltration of bovine serum albumin (BSA). When feed solution flowing perpendicular to the patterns, they observed that the fouling could be mitigated; however, the fouled surface was washed more effectively for parallel feed solution.[14] Using a newly developed layered interfacial polymerization, Choi et al. produced a sharklet-patterned membrane, which exhibited remarkably lower biofouling compared to conventional membranes with irregular roughness and simple
patterns.\cite{15} The sharklet patterned membrane was revealed to possess optimal anti-biofouling effect if the unit and pattern spacings were both 2 μm, as attributed to a balance between intrinsic biofouling propensity and surface flow characteristics such as vortex and primary/secondary flows.\cite{16} Through the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory and density functional theory (DFT) calculations, Li et al. theoretically examined the effect of surface roughness on membrane fouling caused by alginate adhesion. A rough membrane was found to be less alginate adhesion and adhesive fouling than a smooth counterpart.\cite{17}

As discussed above, the fundamental understanding in the effect of surface roughness on membrane fouling is elusive and most of the current studies are focused on polymer membranes. In the past decade, ceramic membranes have been increasingly applied for industrial/municipal wastewater and drinking water treatment.\cite{18} Compared to polymer membranes, ceramic membranes particularly alumina-based exhibit stronger mechanical strength, higher water permeability, better chemical resistance, as well as longer lifetime. Recently, Lyu et al. developed a surface-patterned alumina ceramic membrane with gradient porous structure by using a 3D-printing technology, and the line-patterned membrane exhibited a notable increase in water flux and a significantly enhanced antifouling ability.\cite{19} Alumina membranes with roughness of nm were also reported, e.g., 6.5 nm (Whatman Anodisc) and 2.8 nm (Millipore Nucleopore) for commercial alumina membranes and 4–6 nm for membranes formed by carboxylate–alumoxane nanoparticles.\cite{26} Because the foulant–surface interaction depends on the relative dimension of foulant to surface roughness, the size of foulant is also important. We choose two organic molecules, sucralose (SUC) and bisphenol A (BPA), as illustrated in Figure 1c,d, as foulants. Both foulants are commonly present in wastewater and have a similar size of about 1.2 nm, which is larger than the surface roughness. Following this introduction, the simulation models and methods are briefly described in Section 2. In Section 3, foulant mobility is first examined on the two types of membrane surfaces; then, the effects of surface chemistry and roughness on fouling are discussed in detail. Finally, the concluding remarks are summarized in Section 4.

2. Models and Methods

Figure 1e illustrates a typical simulation system with a foulant molecule solvated in water about 12 Å from an alumina surface. The surface was at the bottom of a simulation cell with a water compartment above and all the surface atoms were fixed. A plate (grey) was above the water compartment to separate it from the vacuum. To avoid the effect of the plate, the water compartment
The model system consisted of a foulant molecule, a monolayer of alumina or graphene, and water. The alumina surface consisted of two layers of aluminum (Al) and oxygen (O) atoms underneath hydrogen (H) atoms. The simulation cell was approximately 4 nm in both x- and y-axes. The alumina surface was considered to be fully hydroxylated with 15 hydroxyl group per nm². Along the z-axis, the alumina surface consisted of two layers of aluminum (Al) and oxygen (O) atoms. The simulation cell was sufficiently large of about 10 nm along the z-axis.

For rough alumina and graphene surfaces, the roughness was introduced onto a flat surface by using a sinusoidal function:

$$z = A \sin \left( \frac{2\pi x}{40} \right)$$

(1)

were A is the wave amplitude, x is the atomic position along the x-axis. For each type of surface, three different amplitudes (A = 0, 3, and 5 Å) were represented by A0, A3, and A5, respectively, and examined. The atoms in alumina and graphene were described by the nonbonded Lennard-Jones (LJ) and electrostatic potentials

$$\sum 4\epsilon_{ij} \left[ \frac{\sigma_{ij}^{12}}{r_{ij}^{12}} - \frac{\sigma_{ij}^6}{r_{ij}^6} \right] + \sum \frac{q_i q_j}{4\pi\varepsilon_0 r_{ij}}$$

(2)

where $\epsilon_{ij}$ and $\sigma_{ij}$ are the potential well and collision diameter, $r_{ij}$ is the distance between atom i and j, $q_i$ and $q_j$ are the charge of atom i and j, and $\varepsilon_0$ is the permittivity of vacuum. The LJ parameters and atomic charges of alumina were adopted from the CLAYFF force field, which is widely used for hydroxylated oxide materials such as alumina, aluminosilicates, and silica. The LJ parameters and atomic charges of graphene were taken from the OPLS-AA force field. The combination of different force fields has been reported in the literature. For example, the CLAYFF force field and AMBER force field were combined to investigate bio-organic materials at mineral interfaces. The OPLS-AA was combined with AMBER force field to predict free energies for ionic liquid-assisted exfoliation of a graphene bilayer. Water was mimicked by the SCP/E model because of its simplicity and capability to reproduce water diffusivity. The LJ and bonded parameters of SUC and BPA were based on the AMBER force field, while their atomic charges were estimated from our previous study. Specifically, SUC and BPA were structurally optimized using the B3LYP functional and 6–311+G(d,p) basis set, then the electrostatic potentials were calculated at 6–311+G(d,p) basis set. The calculations were performed using GAUSSIAN 09 package. Thereafter, the atomic charges of SUC and BPA were estimated by using the restricted electrostatic potential (RESP) method.

Each system was first equilibrated by MD simulation for 100 ps, with a pressure of 1 bar applied on the plate above water compartment. Then, the MD simulation continued for 10 ns with the plate fixed. The simulations were performed at 300 K using velocity-rescaling thermostat and the time step was 2 fs. The electrostatic interactions were calculated by the Particle-Mesh Ewald method, while the LJ interactions were estimated using a cutoff of 1.2 nm. During the 10 ns MD simulation, the foulant molecule was free to move and its mean-squared displacement (MSD) was averaged from the second 5 ns trajectory

$$\text{MSD}(t) = \left\langle |\Delta r(t)|^2 \rightangle$$

(3)

where t is time and $\Delta r(t)$ is the displacement of the foulant molecule. For each system, six independent runs were performed with different random seeds to generate initial velocities.

In addition, the potentials of mean force (PMFs) for foulant on flat alumina and graphene surfaces were calculated separately. The PMF quantifies how the free energy varies when a foulant molecule approaches the surface. To calculate, a force was applied to steer foulant molecule moving toward the surface. Then, umbrella sampling was used involving 21 simulations with 0.05 nm interval along the z-axis. In each simulation, the position of foulant was restrained in the z-axis by a harmonic potential with a force constant of 8000 kJ mol⁻¹ nm⁻². The PMF was then generated using the weighted histogram analysis method. All the simulations in this study were conducted using GROMACS 5.1.4.

3. Results and Discussions

3.1. Foulant Mobility

The MSD of foulant can be used to assess membrane fouling. When a foulant molecule is less mobile, it tends to deposit on the membrane surface and leads to fouling. Figure 2 shows the MSDs of SUC and BPA on alumina and graphene surfaces, respectively, by varying roughness. Each MSD curve is averaged over six independent runs, thus mitigating the effect of random diffusion. There are several interesting observations: (1) on the same surface (e.g. alumina A0), SUC has a smaller MSD than BPA, indicating more severe fouling. (2) On alumina surfaces, both foulants exhibit a similar trend of MSD with roughness; specifically, the MSD rises with increasing roughness (i.e., from A0, A3 to A5). This implies the foulant is more mobile and the fouling is reduced on a rougher alumina surface. (3) The opposite trend is observed on graphene surfaces for both foulants; with increasing roughness, the MSD drops and fouling is enhanced. These observations reveal that fouling is less dependent on the type of foulant, as both SUC and BPA possess similar fouling behavior on alumina or graphene surfaces; instead, the surface chemistry and roughness are dominant factors to determine fouling, which will be discussed in detail below.

3.2. Effect of Surface Chemistry

To quantitatively examine the effect of surface chemistry on fouling, we focus on the flat alumina and graphene surfaces. Figure 3 shows the PMFs of SUC and BPA as a function of distance for the alumina surface. While both foulants show a similar trend of PMF, SUC has a lower minimal PMF (~16.6 kJ mol⁻¹ at point 2) than BPA (~15.6 kJ mol⁻¹ at point 4). The lower PMF of SUC is consistent with the above smaller MSD of SUC on Figure 2a, as a stronger interaction between foulant and surface causes less mobility. For each foulant, the second minimum is observed in the PMF profile (point 1 for SUC and point 3 for BPA). The four points (1, 2, 3, and 4) correspond to different preferred configurations when SUC and BPA are adsorbed on the alumina surface. At point 1, SUC is away from the surface, but comparatively closer to the surface at point 2; consequently, SUC interacts with...
the surface more strongly at point 2 than at point 1, as observed in the PMF profile. For BPA, one of its two ring is parallel to the surface at point 3 and the interaction is stronger than at point 4 without ring parallel to the surface.

On the flat graphene surface, there is distinct difference in the PMFs between SUC and BPA. As shown in Figure 4, the minimal PMF is about $-65$ kJ mol$^{-1}$ for SUC at point 1, while it is $-42.9$ kJ mol$^{-1}$ for BPA at point 3. Meanwhile, the second minimum is seen at point 2 for BPA. The three points (1, 2, and 3) are attributed to different configurations on the graphene surface. From Figures 3 and 4, apparently, the PMFs of both foulants on the graphene surface are much deeper than on the alumina surface. The reason is that the hydrophobic graphene surface interacts more strongly with the two organic foulants compared to the hydrophilic alumina counterpart. Interestingly, as shown in Figure 2, the MSDs of the two foulants appear to be larger on the graphene surface than on the alumina surface. With a closer look at the respective MSDs in the $z$ and $xy$ directions, as shown in Figure S1 in the Supporting Information, we find that the foulant has a negligible MSD, but a significant MSD$_{xy}$. This is attributed to the atomic smooth surface of graphene, thus the foulant can move quite freely parallel to the surface. However, the movement perpendicular to the surface is largely prohibited due to strong interaction. Such a phenomenon was also observed in a simulation study for polymer chains adsorbed on the graphene surface.$^{[42]}$

3.3. Effect of Surface Roughness

Foulant under cross-flow conditions often encounters a shear force which critically affects the fouling behavior of the membrane. Here, we manually add this kind of shear force by adding

![Figure 2](image-url) 

**Figure 2.** MSDs of SUC and BPA on a) alumina and b) graphene surfaces by varying roughness. Each MSD curve is averaged over six independent runs. The shaded region denotes statistical uncertainty.

![Figure 3](image-url) 

**Figure 3.** (Left) PMFs of SUC and BPA on the flat alumina surface as a function of distance. The insert illustrates a foulant molecule moving toward the surface. (Right) Simulation snapshots at different distances (as labelled in the left figure) from the flat alumina surface.
mol\(^{-1}\) with graphene surface; the interaction energies are about principle, the ridge is the place where foulant is most likely to decrease, which causes weaker foulant–surface interaction. In mentally observed. By contrast, the contact area in the ridge comes stronger. This leads to "valley clogging" effect as experimentally observed. From these results, apparently, fouling on alumina surfaces is reduced upon increasing roughness. This prediction is accord with a recent experimental study, in which a 3D-printed line-patterned alumina membrane was observed to be less fouling. To further quantify the effect of roughness on hydrophilic alumina surfaces, we analyze the hydrogen bonds between water and alumina surfaces as illustrated in Figure 7a. Figure 7b shows the numbers of hydrogen bonds on different alumina surfaces. With increasing roughness and hence increasing surface area, more hydrogen bonds are formed between water and the surface. The dynamics of hydrogen bonds can be evaluated by autocorrelation function

\[ c(t) = \frac{\langle h(t_0) h(t_0 + t) \rangle}{\langle h(t_0) h(t_0) \rangle} \]  

where \( h(t) = 1 \) if a hydrogen bond remains at time \( t \) and \( h(t) = 0 \) if otherwise. The ensemble average \( \langle \cdot \rangle \) is on all the hydrogen bonds formed. The \( c(t) \) quantifies the lifetime of hydrogen bonds. From Figure 7c, it is obvious that the \( c(t) \) decays slower on a rougher alumina surface, which implies the hydrogen bonds are more stable and have a longer lifetime with increasing roughness. Thus, roughness increases not only the number of hydrogen bonds, but also the stability. Overall, the surface hydrophilicity of alumina surface is enhanced upon increasing roughness, which is reflected by the interaction energies between water (for all the water molecules) and alumina surfaces in Figure 7d.

It is instructive to explore how the fouling behavior on hydrophilic alumina surfaces changes to that on hydrophobic graphene counterparts. To achieve this, we manipulate the hydrophilicity of alumina surfaces by scaling the atomic charges of

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**Figure 4.** (Left) PMFs of SUC and BPA on the flat graphene surface as a function of distance. The insert illustrates a foulant molecule moving toward the surface. (Right) Simulation snapshots at different distances (as labelled in the left figure) from the flat graphene surface.

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**Figure 6b and Figure S4b (Supporting Information)** shows the contact areas between BPA and alumina surfaces. As also observed on graphene surfaces, the contact area increases in the valley and decreases in the ridge. On alumina surface A5, the contact area may become 0, which implies the foulant leaves the surface and the foulant–surface interaction decreases to 0 accordingly. From these results, apparently, fouling on alumina surfaces is reduced upon increasing roughness. This prediction is accord with a recent experimental study, in which a 3D-printed line-patterned alumina membrane was observed to be less fouling. To further quantify the effect of roughness on hydrophilic alumina surfaces, we analyze the hydrogen bonds between water and alumina surfaces as illustrated in Figure 7a. Figure 7b shows the numbers of hydrogen bonds on different alumina surfaces. With increasing roughness and hence increasing surface area, more hydrogen bonds are formed between water and the surface. The dynamics of hydrogen bonds can be evaluated by autocorrelation function

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Figure 5. a) SUC moving along graphene surfaces under a harmonic force in the x-axis. b,c) Interaction energies and contact areas between SUC and graphene surfaces.

Figure 6. a) BPA moving along alumina surfaces under a harmonic force in the x-axis. b,c) Interaction energies and contact areas between BPA and alumina surfaces.
alumina. For example, if a scaling factor is 0.5, the atomic charges are only 0.5 times of the actual alumina. Such a simple scaling method was used in the literature to change the hydrophilicity of MgO surface. Figure S7 (Supporting Information) shows the interaction energies between water and alumina surfaces with different scaling factors. By decreasing the scaling factor, the water-surface interaction becomes weaker and apparently the surface is less hydrophilic. Figure 8 shows the MSDs of BPA on alumina surfaces with different scaling factors. On the actual alumina surfaces (i.e., scaling factor = 1), as shown in Figure 2a, the MSD rises upon increasing roughness and hence fouling is reduced. When the scaling factor = 0.7, the MSD is nearly equal on A5 and A0 surfaces. When the scaling factor = 0.1, at which the surface is highly hydrophobic, the trend of MSD resembles that on graphene surfaces in Figure 2b; i.e., the MSD drops on a rougher surface and fouling is enhanced. Clearly, these results reveal that fouling behavior can be tailored by changing the hydrophilicity or hydrophobicity of surfaces. This further suggests that a less rough surface is prone to reduce fouling on a hydrophobic surface like graphene. On a hydrophilic surface like alumina, however, fouling is reduced upon increasing roughness.

4. Conclusions

We have conducted molecular simulations to investigate the effects of surface chemistry and roughness on membrane fouling. For two foulants (sucralose and bisphenol A), the mobility on alumina surfaces is found to rise with increasing roughness and hence fouling is reduced by roughness. The opposite trend is observed on graphene surfaces, i.e., the mobility of foulant drops and fouling is enhanced with increasing roughness. These interesting opposite effects of roughness on fouling are attributed to the different foulant-surface interactions and contact areas between alumina and graphene surfaces. It is also found that upon increasing roughness, more hydrogen bonds with longer lifetime are formed between water and alumina surface. Furthermore, the fouling behavior can be readily tuned by varying the hydrophilicity/hydrophobicity of surfaces. The microscopic insights into the crucial effects of surface chemistry and roughness on fouling from this bottom-up study are useful for the development of antifouling membranes for water treatment and other important separation processes.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

The authors gratefully acknowledge the Singapore National Research Foundation (NRF-CRP17-2017-01), the Singapore Ministry of Education and the National University of Singapore (R-279-000-598-114, R-279-000-574-114, and C-261-000-207-532/C-261-000-777-532) for financial support.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.
Keywords
alumina, fouling, graphene, roughness, surface chemistry

Received: September 16, 2021
Revised: October 14, 2021
Published online: November 20, 2021

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