Crumpled Graphene Oxide-Polysulfone Composite Membranes for Water Purification

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Abstract:

Previously, crumpled graphene oxide (CGO) has been layered on surfaces of commercial water treatment membranes to increase water flux as well as antimicrobial (anti-biofouling) properties. This study explored CGO's effects on flux, rejection, fouling, and antifouling properties when structurally incorporated into polysulfone (PSF) membranes. Different ratios of CGO were added into the casting solution for a phase-inversion process. The synthesized membranes were then tested in an effort to optimize CGO mass loadings. Proper methods and variables were found and accounted for throughout the series of experiments. Flux and rejection were found as high as 100 L/m² h bar and 90% of BSA respectively. It was hypothesized that surface openings from CGO could serve as water channels through the selective surface toward the more porous region of the membrane, creating higher flux.

Introduction:

Current water treatment methods employ hollow-fiber ultrafiltration (UF) membranes in vast quantities to filter macromolecular impurities and bacteria from feed water for various industrial and residential applications. These modules of membranes require expensive and time-consuming cleaning processes, such as backwashing, in order to remove residue buildup on membrane surfaces. Given the hydrophilic properties of graphene oxide (GO), some research for advancements in water treatment membranes has been geared towards GO nanoparticles.

Previous research has shown that crumpled graphene oxide (CGO) structures can increase flux due to its hydrophilic properties and porous nature [1]. In this study, the effects on polysulfone membrane properties were investigated extensively to determine the performance of CGO as a nanofiller to membrane casting solutions prior to solidification.

Experimental:

Membranes were created using the phase-inversion method [2]. CGO (5-10 mg) was sonicated in n-methyl-2-pyrrolidone (NMP), then stirred with polysulfone (PSF) and polyvinylpyrrolidone (PVP) at 60°C until becoming homogenous. The solution was cast using a casting knife and then submerged in MilliQ water overnight. Subsequently, 63 mm diameter membranes were cut with razor blades.

Transmission electron microscopy (TEM, Tecnai TM Spirit) was utilized to obtain information on CGO and CGO nanocomposites' size and morphologies. Scanning electron microscopy (SEM) images of the membranes (sputtered with Au for 60 s) were taken to examine surface structure.

Membranes were then tested for flux by filtering pure water with a stirred cell under 1.5 bar (Amicon 8200) and measuring permeate weight over 60 s time intervals. Membranes were compacted under 1.5 bar for one hour before any measurement. Water was filtered through the membrane until flux plateaued. Bovine serum albumin (BSA) (1 g/L) was filtered separately as a model foulant. Rejection was determined by the difference between inflow and permeate BSA concentrations that were analyzed at 278 nm using UV-visible spectrophotometry (UV-Vis). All samples were washed with water for several minutes before storing in pure water. Filtration processes were repeated to obtain consistent data as well as fouling and antifouling properties.

Results and Conclusions:

SEM and TEM images in Figure 1 and Figure 2 of CGO membranes show embedded CGO particles directly underneath and protruding from the surface of the membrane. Pristine (control) membranes showed smooth surfaces without notable inconsistencies, forming a smooth solid barrier. CGO nanoparticles channel water from the surface directly into the porous region of the membrane. These particles could be acting as hydrophilic pathways with lower resistance and higher localized flux

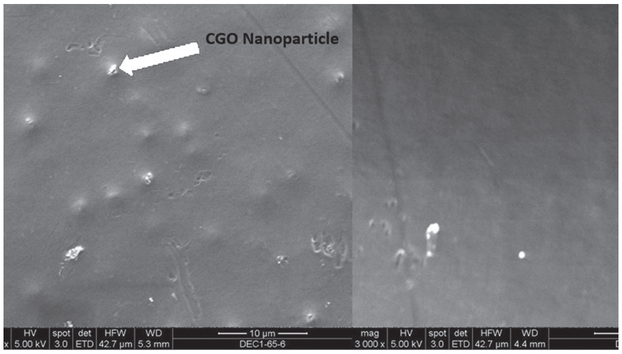

Figure 1: SEM images of CGO nanoparticles protruding through surface of CGO membrane (left) and surface of pristine membrane (right).

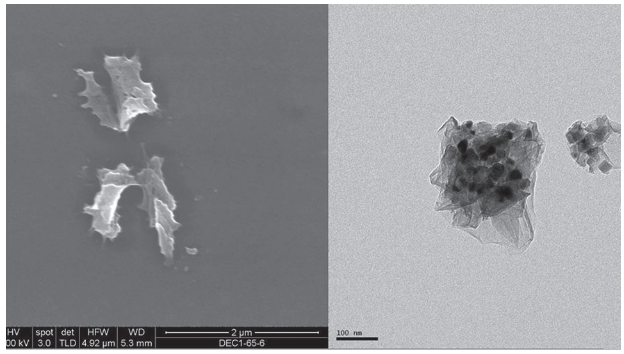

Figure 2: SEM of embedded CGO particle (left) and TEM of CGO with titanium and $CuCl_2$ from aerosolized process [3] (right).

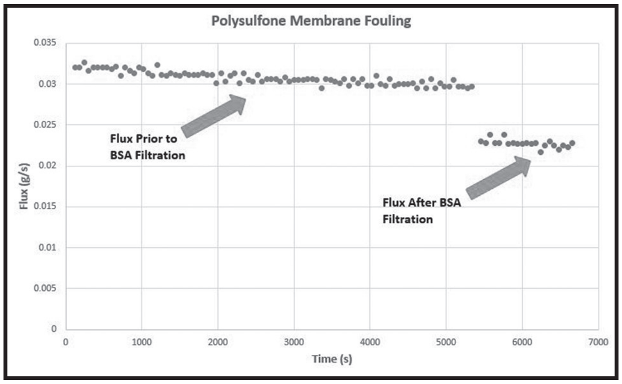

Figure 3: Control membrane fouling before and after BSA filtration.

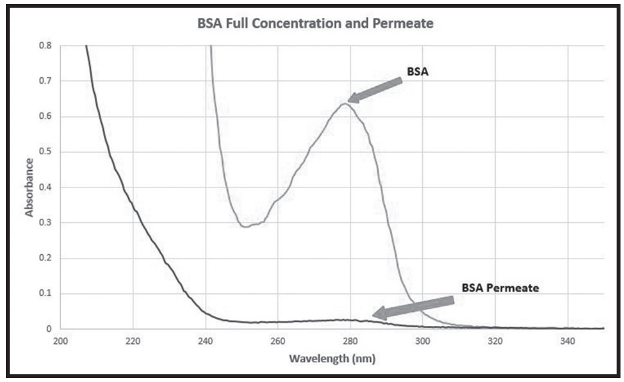

Figure 4: UV-Vis spectrum for inflow BSA and permeate used to determine rejection.

than the surrounding surface, resulting in larger overall flux than membranes without such channels.

Figure 3 shows the fouling process for a pristine membrane. Water flux was found to plateau until a constant value of 0.0295 g/s was reached. After BSA was filtered through the membrane, water flux decreased to 0.0226 g/s — 76.6% of the initial value. Foulant built up on the membrane surface, obstructing pores and decreasing flux. Despite a washing process, not all foulant was removed, damaging the membrane and resulting in a permanently lower flux. This pattern was consistent for both control and CGO membranes alike.

During UV-Vis tests for BSA, absorbance peaked at a wavelength of 278 nm. The linear relationship between concentration and absorbance was compared with the BSA permeate absorbance of each membrane to obtain the remaining BSA in the permeate. An example of a control membrane is given in Figure 4, which has the inflow solution peaking at 0.616 and the permeate at 0.0238, indicating a 96.1% rejection for the given filtration trial.

Rejection for control and CGO membranes ranged from 73.9-96.1% and 70.9-99.2%, respectively, while flux ranged from 0.0176-0.0307 g/s and 0.0155-0.0317 g/s, respectively. Methods to measure both fouling and rejection were refined and will be used for future experiments. Though no statistical differences between CGO and control membranes were found, our preliminary results give base data and processes that will be built further upon by continued research.

Future Work:

The largest issue with results was variability within casting solutions. In future studies the focus should be on consistency and repeatability at each step of experimental processes. Strong evidence was found to support that without the drying process, membranes had up to 10 times higher water flux than dried samples of the same composition, but with a drop of rejection as low as much as 50%. This relationship should be investigated to obtain optimum performance.

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