



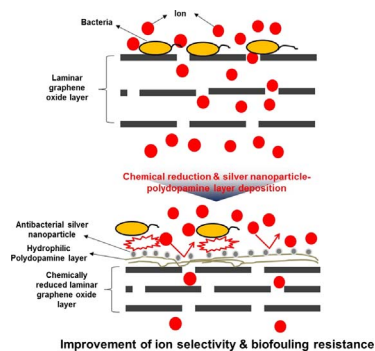
Laminar reduced graphene oxide membrane modified with silver nanoparticle-polydopamine for water/ion separation and biofouling resistance enhancement



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GRAPHICAL ABSTRACT



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ABSTRACT

Laminar graphene oxide (GO) membranes with excellent water permeability have demonstrated great potential in the field of membrane-based water purification. However, low ion rejection and controversy over their biofouling propensity are hurdles GO membranes face in realizing commercial applications. This study shows that silver nanoparticle (nAg)@polydopamine (pDA) deposition on chemically reduced GO (rGO) laminates can simultaneously enhance both biofouling resistance and ion rejection. In pressurized filtration tests, the salt rejection of nAg@pDA-rGO membranes outperformed that of GO membranes despite its lowered water permeability. In osmotic filtration tests, nAg@pDA-rGO membranes achieved an increased water flux of 28.9 LMH and reduced reverse solute flux of 0.21 mol/m²h, compared to GO membranes. Moreover, extremely low cell attachment and viability were seen on the nAg@pDA-rGO membrane, confirming the excellent antibiofouling ability of nAg-pDA-rGO membranes.

1. Introduction

Membrane-based water purification technologies are being highly regarded as the most reliable way to guarantee the quality of global potable water supplies. However, membrane-related challenges such as

fouling and the relatively high energy requirement compared to conventional water production technologies remain unresolved, caused by the performance limitations of current polymeric membranes [1,2]; innovation in membrane materials and fabrication is required in order to resolve these membrane-related problems.

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The development of next-generation water purification membranes using graphene oxide (GO) nanosheets is a vibrant and active research field [3,4]. GO nanosheets are graphene-based nanoplatelets that include diverse oxygen functional groups (e.g., carbonyl, carboxyl, epoxy, and hydroxyl) [5]. GO nanosheets have definite merits as water purification membrane materials, such as high hydrophilicity, negative surface charge, atomic-like thickness, robust mechanical strength, and are chemically stable [3]. To fabricate water purification membranes using GO nanosheets, the sheets are stacked using various methods (e.g., vacuum-assisted filtration [6], layer-by-layer [7], and spin-coating [8]); nanochannels are subsequently formed in the stacked-GO layer. These nanochannels play significant roles in facilitating fast water transport passage and as a selective molecular barrier in GO membranes [9]. In a previous study [7], GO membranes achieved a maximum 10 times higher water permeability and a higher rejection rate for ~500 Da organic dye molecules than commercial nanofiltration membranes.

However, critical challenges such as the instability of GO laminates in aqueous solution, poor monovalent ion rejection rate, and a dispute over the anti-microbial properties of GO membranes remain [10–12]: First, extremely hydrophilic GO nanosheets can be easily hydrated, such that the structure of the GO laminates loosens, as GO nanosheets consisting of GO laminates eventually re-disperse into the aqueous solution [12]. Second, despite the excellent rejection rate toward molecules and large ionic species, GO membranes could not achieve a high rejection rate for small ions [6,7]. To reject monovalent ions, theoretically, the size of GO nanochannels needs to be less than 7 Å because the radius of hydrated sodium ions is 3.6 Å [9]. The GO laminates loosened by the effect of hydration generally have ~9 Å nanochannels; therefore, small monovalent ions can easily penetrate through the GO nanochannels [10]. Third, in terms of the antibacterial effect of GO membranes, several conflicting results have been reported. For example, Das et al. [13] showed that there was no growth inhibition zone around GO film. In another study, Ruiz et al. [11] demonstrated that GO membranes had no antibacterial effect, but rather promoted bacterial growth. In contrast, Hu et al. [14] demonstrated GO and rGO membranes fabricated via vacuum filtration had the excellent bactericidal effect. Similarly, Bao et al. [15] exhibited GO papers can inhibit growth of *E. coli* and *S. aureus* bacterial cells. More recently, Perreaut et al. [16] reported that GO membranes have quite strong antimicrobial properties, although they depends on the GO nanosheet size.

These bottlenecks must be eliminated prior to their practical application in water/ion separation systems. However, to the best of our knowledge, there have hitherto been only few studies on fouling propensity of laminar GO membranes for water/ion separation [17,18]. Sun et al. [17] fabricated laminar membranes by stacking silver nanoparticle (nAg) decorated GO nanosheets via a vacuum-assisted filtration method, and evaluated performance and biofouling propensity. The nAg-decorated GO membranes exhibited fabulous water flux and anti-biofouling capability, but the membrane might not be suitable for ion rejection; the size range of the nAgs on GO nanosheets was from 5 nm to 15 nm, implying that the nanochannel size of nAg-decorated GO membranes may be too large to reject ions. In another study conducted by Hu et al. [18], fouling resistance and membrane performance of laminar GO membranes fabricated by alternatively assembling GO nanosheets and poly(allylamine hydrochloride) via a layer-by-layer method were assessed in an osmotically-driven system. They showed the GO membranes showed comparable organic fouling propensity to commercial polyamide membranes. However, biofouling propensity of the GO membrane was not evaluated.

Therefore, we have attempted to evaluate biofouling resistance of GO-based membranes (e.g. GO and reduced-GO (rGO) laminates) and develop laminar GO membranes capable of rejecting ions and highly inhibiting biofouling by employing three simple and easy technological approaches. First, GO laminates fabricated via a vacuum-driven filtration method are reduced using hydroiodic acid vapor to narrow the GO

nanochannels. The hydroiodic acid vapor reduction eliminates oxygen functional groups in the GO nanochannels subsequently, the nanochannel become narrow. The narrowed nanochannels can reject more ions. Second, a layer of hydrophilic polydopamine (pDA), a bio-mimetic substance inspired from a mussel adhesive foot protein [19], is applied as a coating on rGO laminates by dipping in an alkaline dopamine solution. In an oxygen-rich alkaline condition, catechols in dopamine are oxidized to quinones thereafter, the quinones react with other quinones or catechols for pDA formation [20]. Due to plentiful catechol moieties in pDA, the pDA coating layer has a strong adhesive force and high hydrophilicity [21,22], which can produce mechanically strong and hydrophilic membrane surface. The hydrophilic surface could improve water permeation and mitigate bacterial attachment. Moreover, catechol functional groups in the pDA layer can reduce metal ions to nanoparticles [23]. Finally, nAgs, which has a superior bactericidal effect but a low toxicity to human cells [24], are deposited on pDA-coated rGO laminates by reducing silver ions through catechol functional groups, which have the ability to reduce metal ions [23,25–27]. The nAg deposited on the pDA coating layer can enhance biofouling resistance.

Note that the physicochemical properties of nAg-pDA-coated rGO (nAg@pDA-rGO) membranes are first compared to those of GO, rGO, pDA-rGO membranes. Performance of the nAg@pDA-rGO membranes are then evaluated in both pressurized filtration and forward osmosis (FO) systems; it has been reported that ion and water permeation across laminar GO membranes under osmotically-driven conditions is quite different from that under hydraulic pressure-driven conditions [28,29]. The biofouling resistance of the nAg@pDA-rGO membranes is subsequently confirmed through bacterial attachment, zone of inhibition tests, and biofouling tests in a crossflow FO system.

2. Materials and methods

2.1. Fabrication of nAg@pDA-rGO membranes

A high concentration GO solution (6.2 mg/mL), manufactured by Graphene Supermarket (USA), was first pretreated by the following steps: 1) sonication for 90 min, 2) dilution to 1 mg/mL, 3) centrifugation at 10,000 rpm for 1 h, 4) disposal of precipitate, and 5) sonication for 90 min. The pretreated GO solution was then diluted with deionized water to make a 0.006 mg/mL high-purity and homogenous GO solution. Next, 50 mL of the as-prepared GO solution (0.006 mg/mL) was vacuum-filtered through a 0.2 μm mixed cellulose ester membrane (MCE; diameter: 47 mm; Adventec MFS Inc., Japan) to synthesize a stacked-GO membrane (Fig. 1). After drying the as-prepared laminar GO membranes at a room temperature for 24 h, the laminar GO layer of the resultant membranes was chemically reduced using hydroiodic acid (HI; Sigma Aldrich, USA) steam. The rGO membranes were then placed in a desiccator at room temperature for 24 h to remove residual hydroiodic acid from the membranes. A pDA layer was deposited onto the rGO membranes by immersing them in an oxygen-rich dopamine solution (2 g/L, pH 8.5), prepared by dissolving dopamine hydrochloride (Sigma Aldrich, USA) in a Tris-HCl buffer solution (15 mM, pH 8.5), for 1 h. The resultant pDA-rGO membranes were desiccated, before being submerged in a 0.5 M silver nitrate solution, which was prepared by dissolving silver nitrate powder (Junsei Chemical Co., Ltd., Japan), for 12 h under UV irradiation to coat the nAg onto the pDA-rGO membranes. The power of UV light source used in this work was 4 W, and the distance between the UV light source and sample was about 40 cm.

To compare the as-prepared laminar GO membranes, commercial cellulose triacetate embedded support FO membranes (CTA ES, Hydration Technology Innovations, USA) were prepared. The properties of the CTA ES membranes are provided in Supplementary information (Fig. S1 and Table S1).

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