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Review

Review on structural control and modification of graphene oxide-based membranes in water treatment: From separation performance to robust operation☆

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ABSTRACT

Membrane separation has become an important technology to deal with the global water crisis. The polymer-based membrane technology is currently in the forefront of water purification and desalination but is plagued with some bottlenecks. Laminated graphene oxide (GO) membranes exhibit excellent advantages in water purification and desalination due to the single atomic layer structure, hydrophilic property, rich oxygen-containing groups for modification, mechanical and chemical robust, anti-fouling properties, facile and large-scale production, *etc.* Thus the GO-based membrane technology is believed to offer huge opportunities for efficient and practical water treatment. This review systematically summarizes the current progress on the water flux and selectivity intensification, stability improvement, anti-fouling and anti-biofouling ability enhancement by structural control and modification. To improve the performance of the laminated GO membrane, interlayer spacing tunability and surface modification are mainly used to enhance its water flux and selectivity. It is found that the stability and biofouling also block the service life of the GO membrane. The crosslinking method is found to effectively solve the stability of GO membrane in aqueous environment. Introducing nanoparticles is a widely used method to improve the membrane biofouling ability. Overall, we believe that this review could provide benefit to researchers in the area of GO-based membrane technology for water treatment.

1. Introduction

The increasing population growth, urbanization and industrialization have led to the growing water consumption and uncontrollable wastewater discharge [1,2]. In order to address the global water crisis, membranes for water purification and desalination are becoming more and more popular due to the its advantages of low energy consumption, low investment cost, ease of operation and possibility for continuous operation [3,4]. High selectivity and permeability are the key properties [illegible]. A tremendous amount of effo[illegible] and in[illegible]

The c[illegible] be classifie[illegible] Undoubtedly,

desalination and wastewater treatment due to their excellent cost-effective and defect-free production. Inorganic membranes have excellent anti-pollution ability, high tunability and reusability, and good chemical stability [6,7]. The organic/inorganic membranes combine the unique properties of both polymeric matrix and inorganic filters, making a great progress in dealing with the critical issue of trade-off relationship between permeability and electivity as well as membrane fouling and scaling [8,9]. One of the most remarkable achievements is the development of mixed matrix membranes (MMMs) and thin film composite (TFC) membranes [10,11].

The recent rise of nanotechnology has opened up a new way to [illegible] materials into the field of membrane [illegible]

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Fig. 3. Comparisons of the structure and stability of the common GO and the proposed GO/prGO

increase the permeability and selectivity of the composite GO membranes, which can be achieved by controlling proper concentration of OCNTs in the GO suspension during the membrane fabrication.

cently, the GO interlayer spacings were successfully adjusted and y intercalating cations into the GO interlayers [52]. The lam- brane was immersed in the saline solution, and then the bed into the GO interlayers. The successful intercala- confirmed by molecular dynamics (MD) simula- l theory (DFT) calculation. On one hand, it hat the hydrated cations could bind the GO sheets through a stable hydro hand, DFT calculations indicated that groups and the aromatic rings coexist pro adsorbing the cations. Therefore, the concen tion determines the interlayer spacings of the film. The GO interlayer spacings were fixed by the interaction including the cation–π interaction betwe cations and the basal plane of the GO nanosheets, as well a tion between the hydrated cations and the oxygen-containi on the GO sheet.

2.2. Graft by functional groups

Sulfuric acid was adopted to graft sulfonate group with high hydrophilicity on the surface of GO sheets [53,54]. The sulfonated GO nanosheets are endowed with enhanced hydrophilicity. The sulfonic groups on the GO surface have strong hydrogen bonding interaction with water, thus producing a layer of water molecules on the membrane surface. It was claimed that reasonable sulfonation could improve the water permeability of the laminated GO membranes, and the water permeability increases with the increasing the sulfonation degree. Nevertheless, excessive sulfonation has negative impact on the improvement of water permeation through the laminated GO membranes.

To improve the surface property of the GO nanosheets, Kochameshki *et al.* [46] grafted hydrophilic/charged polymer chains on tTdreth gc2.631-1.316Tdplane5(s-50933(I314(t))-13(w13(as508)3(r)-5(e)poraft)-5das

sheets [56].

The crosslinked GO
as rinsed in water
olecules, such as
DA) [56], hex

Fig. 11. Schematic of silver nanoparticle interaction with bacterial cells [75].

(such as anti-adhesion, shape puncture through the cytomembrane, and oxidation of cellular components) as well as the electrostatic interaction between the bacterial cytomembrane and the nanoparticles or chitosan. As shown in Fig. 12, compared with the control image clearly, biofilm could not be observed on the surface of the four nanocomposite films.

Furthermore, GO functionalized with Ag (Ag-GO) was adopted as the surface coating to enhance the anti-biofouling ability of the laminated GO layers [83]. The Ag-GO coating layer was stably supported on the GO membrane surface. The anchored AgNPs could lower the surface adhesion to bacteria and inactivate the adhered bacteria. Besides, the Ag-GO coating endowed the GO membrane with more smooth and negative zeta potential surface than the pristine GO-stack membrane.

Shuai *et al.* [81] also intercalated AgNPs into the interlayers of the laminated GO layered, and achieved a layer-by-layer structure with

Fig. 12. FESEM images of *E. coli* bacteria attached on (A) control surface, (B) GOns-Ch film, (C) GOns-AgNP film, (D) Ch-AgNP film and (E) GOns-Ch-AgNP film [82].

Fig hematic c he p repa of the co-c GO n and Ag nanoparticles [81].

GO nanosh eets an supp , to each , as shown i g 13. As the bacteria ap to th ace of th -Ag membra t will suffer fro cellul ure d ction an nction d mage t as found tha he an l pro y mainly sulted fro the y al damage o he sh ture e sharp edges int the b ct al cytomem ane a emic tack by A $^{+}$ and RO S.

Faria *et al.* [84] zed cteriostatic layer of Ag s on he face of TF membr *in-situ* ucleation and growt gNPs o e top GO l r. The bio ing tests showed th t *in-situ* duced G Ag lay ted od anti- ofouling a Durin tests, a dead we observe n the far s the bi from th GO n s ace. It dicates th t im membr could act a in ind contact w t me surface s is d act at the sed Ag^{+} io s d f o the su ding t s en he modified me b e with n ontac ob l abi

4.2. D with p rti

Ki *et al.* [8 anocomp it se he bacte ic c t he impro d m ne show t ba s ated and v s, as sh Fig ef (RI) distri ic ea el in th ial et omograp I hic . It in cha acte GO-MoS_2 apopte er 90 n des also O-MoS_2 c roduce R (*i.e.* xid n, O_2 s the ox ve stress o bac tra ar co h as pro s, phosp n and a . The o he intra ular omp lea e d age o cytome rane. Accor the wi severe y omembr es hen lose a Fig ws collecton cells were illed after 3 h for in

structu ibacterial abil nanocom os 5].

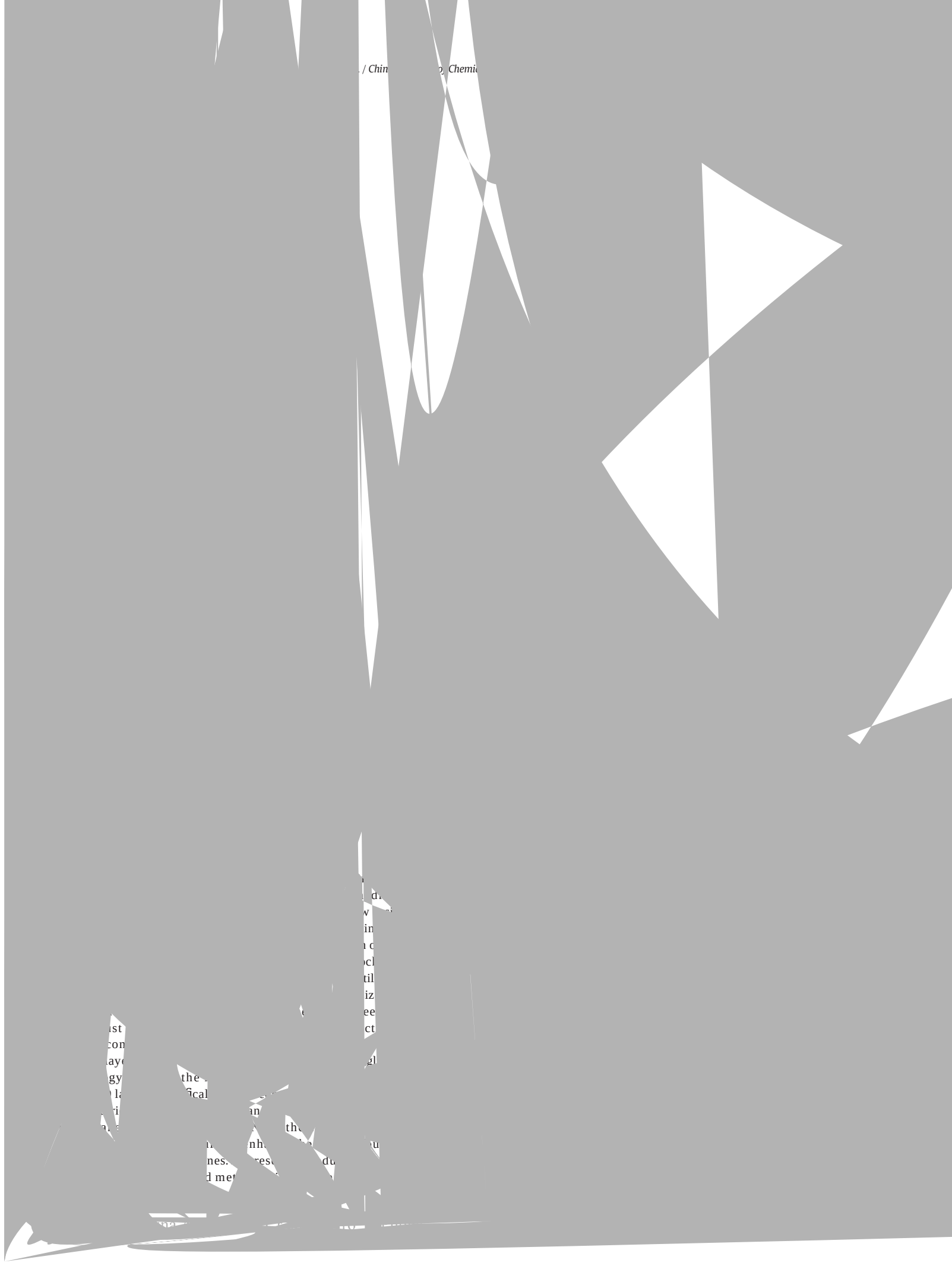
t properties of hydrophilicity, antibacterial, anti- o on nd mechanical stability, thus it is a good additive r al t he anti-fouling and anti-biofouling ability of mem- b e [86] the nano-sized ZnO particles (ZnONPs) are apt to ag egate hus they cannot be well dispersed in the polymer m trix. Re ung *et al.* [87] prepared ZnO-GO nanohybrid *via* the sol–gel confine the ZnONPs aggregation, which improved the dispers ONPs in the membrane. Similarly, ZnONPs could nerate RO disrupts the bacterial cell integrity. Thus the for- tion of t al cells could be suppressed by the presence of ONPs.

Surface n n by chitosan

GO compo osheets were also prepared by surface modifica- of with c 88–91]. Condensation reaction between the car- l group o the amine group of chitosan occurs to form the e covalei which stabilizes the hydrophilicity of the GO brane su 15 depicts the schematic process of synthesizing O nanosh dified by chitosan. The grafted chitosan enhances dr ophilic smoothness of the membrane surface. This is the to improve i-biofouling property of the GO membrane. Fur- re, the p ly charged —NH_2 of the chitosan chains could e ectrostat raction with the anionic groups of the microbial mbrane. ectrostatic interaction weakens the cytomem- l rmeabil even the destruct the cytomembrane, resulting i osis of th robial cells.

5. sions

ne oxic novel material with many advantages that en- abl minate membrane with excellent properties for wate p ification desali tion. A fundamental understanding of t s cture–p y rela ps is critical to effectively improve



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