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Graphene Oxide for Adsorptive Ultrafiltration Membranes

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ABSTRACT

Adsorptive ultrafiltration membrane is an emerging field that holds great promise for achieving high efficiency removal of various pollutants such as metal ions and organic pollutants. As a result, significant research efforts have been directed towards the utilization of GO due to its unique properties including good hydrophilicity, large surface area, excellent chemical and mechanical stability which can impart better membrane properties and performance. Recent advances in this field revealed that the utilization of graphene oxide (GO) is highly beneficial in improving key membrane properties including surface charge, hydrophilicity, water permeability as well as the adsorption performance. Therefore, this short review provides valuable insight into the performance of GO/GO modified-based membranes, GO/polymeric-based membrane and GO-modified/polymeric-based membrane for various pollutants. The application of GO in ultrafiltration (UF) membranes is anticipated to provide an attractive and viable route towards achieving high performance adsorptive UF membranes for water and wastewater treatment.

Keywords: Graphene oxide, adsorptive membrane, ultrafiltration, ions; heavy metals

1.0 INTRODUCTION

Adsorptive ultrafiltration (UF) membrane has emerged as an excellent candidate for wide ranges of applications particularly for water and wastewater treatment process. The technique which involves one-step treatment offers excellent reusability, economic viability, ease of operation as well as high efficiency for pollutants removal even at very low concentration [1, 2]. For instance, incorporation of various adsorptive materials (i.e., nanoparticles) in the membrane matrix endows the mixed matrix membranes (MMM) with excellent adsorptive properties towards various pollutants removal which can be associated with the synergistic properties of adsorptive materials and the membrane matrix [3]. In addition, the desired membrane properties can be easily tuned by varying the membrane material, membrane fabrication conditions and the types of materials introduced in the membrane matrix.

To date, a number of exciting studies have evidenced the versatility and outstanding properties of graphene oxide (GO), a chemical derivative of graphene with various oxygen functionalities (i.e., carboxyl, hydroxyl, epoxy) in the area of adsorptive membrane. One of the key benefits of GO also relies on its ease of functionalization through covalent and non-covalent bonding which can be tailored for removal of specific contaminants in water [4, 5, 6. Moreover, the amphiphilic nature of GO due to presence of oxygen groups

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provides good hydrophilicity while the non-oxidized part acts as a pure graphene, which is beneficial during the membrane filtration process [3]. The outstanding attributes of GO also include exceptional transport properties, high surface area, high affinity, surface with negative charge, desired functional groups and good mechanical strength. Recent studies have also shown that the presence of GO is beneficial in improving anti-fouling properties of UF membranes [4, 7]. The versatility of GO also relies on its nanochannel properties which can promote the selective permeability for outstanding removal and adsorption of various pollutants [8, 9, 10].

Significant studies have unveiled the outstanding potential of GO-based membranes for the removal and adsorption of various pollutants which include dyes, heavy metals and salts. Thus, this review outlines the latest studies on GO-based membranes including reduced GO (rGO) membranes. Moreover, this review also summarizes the latest studies related to GO/polymer-based membranes to provide better understanding of this field, as listed in Table 1. This short review will be of great interest to researcher in the field of water and wastewater treatment who are interested in exploring the potential of GO-based or GO/polymer-based membranes for the removal of different types of contaminants.

2.0 GRAPHENE OXIDE-BASED MEMBRANES

The unique properties of GO such as well-defined nanometer pores, selective permeability, tuneable sieving and exceptional transport properties have made GO-based membrane highly attractive for adsorption of various pollutants. Thus, various efforts have been made to impart better understanding particularly on the roles of GO for the preparation of adsorptive membrane [11, 12, 13]. It is worth noting that the primary advantage of utilizing GO-based membranes is associated with the fast transport of water through the interconnected nanochannels between the adjacent GO nanosheets [7]. However, the application of GO-based membranes is restricted due to several drawbacks such as low selectivity, permeability and stability [14]. According to reports, the use of GO membranes is hindered due their low stability in aqueous solution, primarily due to the electrostatic repulsion of GO nanosheets [14, 15, 16]. This instability could lead to a reduction in membrane performance as well as practical applications. Additionally, another challenge associated with GO membranes is their intrinsic hydrophilicity, which can cause swelling and potentially lead to delamination of the membranes [17. Thus, various modification strategies have been proposed to address the underlying issues. These strategies include covalent and non-covalent crosslinking methods which can enhance the stability and sieving property of GO membranes [15,18]. Other strategies also focus on the development of composite GO membranes by combining GO with other materials (i.e., nanomaterials) to impart better permeability and selectivity [14, 15]. Recently, Ali *et al*. [17] prepared PEGylated GO (PGO)-based membranes by modification of GO lamellar membrane. The membranes with controlled pore sizes were functionalized with 6-armed poly(ethylene oxide) via a simple amidation route.

Table 1 Overview on selected recent researches related to GO adsorptive membranes for various applications

Membrane / main additive	Pollutants adsorption / PWF	Remark	References
GO / CNT	Co(II) = 37 mg g^{-1} Ni(II) = 40 mg g^{-1} Cu(II) = 50 mg g^{-1} Zn(II) = 42 mg g^{-1} Cd(II) = 98 mg g^{-1}	Significant reduction of adsorption capacities was observed with the addition of CNTs.	[18]
GO / *β*-CD)	BPA adsorption = 82 – 166 mg m^{-2} PWF = 837 L/m^2 hr	The adsorption capacity was influenced by the grafting density of *β*-CD, surface area and nanochannel properties.	[29]
GO-PACl	Humic acid = 99% TOC = 90-95% PWF = 130 L/ m^2 hr	Higher water flux and organic molecules rejection can be achieved by varying the OH/Al ratio.	[24]
PSf / GO	Ofloxacin = 90% Benzophenone-3 = 90% Rhodamine = 90% Dichlofenac = 90% triton x-100 = 90%	Loading of GO significantly influenced the performance of the prepared membranes.	[32]
PSf / GO	Lead(II) = 90% Pb^{2+} = 80%	Adsorption capacity was greatly influenced by the pH of solution.	[36]
PVP / GO / PSf	Pb^{2+} : 80% PWF = 150 L/m^2 hr	Addition of PVP enhanced the distribution of GO within the membrane surface.	[38]
PES / GO	Acid black (AB) and rose bengal dyes = 99% PWF = 116 L/m^2 hr	Higher loading of GO led to pore blockage which decreased the membrane permeability.	[31]
GO-PDA / PES-SPES	Rhodamine B = 87 mg g^{-1}	Enhanced adsorption capacities were associated with the synergistic effect between GO and PDA groups.	[4]
PES / QSiPD-rGO	BSA = 98% PWF = 270 L/m^2.hr	Loading of QSiPD-rGO significantly influenced the membrane porosity and permeability.	[40]
GO-UiO-66 / PES	PWF = 16 kg / m^2.hr	Higher loading of GO-UiO-66 could lead to partial agglomeration, decreasing the membrane permeability.	[44]
nano-GO / PSf	PWF = 219 L/m^2.hr	Presence of nano-GO enhanced the pore structure, hydrophilicity, permeability and anti-fouling properties.	[10]

The resulting modified membranes demonstrated an outstanding rejection of Pb^{2+} and Ni^{2+} salts (>80%), 99% rejection of NaCl and enhanced stability. Detailed discussion on GO lamellar membrane can be found elsewhere [19]. Moreover, study by Marcin Musielak *et al*. [18] has proposed an alternative to overcome this stability issue by introducing an oxidized carbon nano tubes (CNTs) into GO-based membranes [18]. The non-covalent interaction between GO and oxidized CNTs has resulted in enhanced membrane durability. However, it is noted the stabilization of the membrane by CNTs has jeopardized the adsorption capacities for the Co(II), Ni(II), Cu(II), Zn(II),Cd(II) and Pb(II). This is due to the formation of interconnected micro and nano-channels from the entangled CNTs. On the other hand, Zhang *et al*. [20] revealed that incorporating isophorone diisocyanate (IPDI) that is cross-linked to GO into GO framework (GOF) membranes resulted in the enlargement of the nano-channels of the GO nanosheets, leading to increased of water flux (80 – 100 L/m^2 hr bar). The GOF membranes also demonstrated excellent removal (>96%) of dyes (methylene blue, methylene orange, rhodamine-B and congo red) [20].

Recent advancement in the utilization of GO adsorptive membranes have also contributed to the understanding from the interlaying spacing perspective [21, 22]. Previous studies have inferred that the interlaying spacing is very crucial in controlling the mass transport behaviour of ions across the membranes [21, 22]. Thus, a promising and efficient technique of controlling the interlaying spacing of GO nanosheets is highly desirable. Tan *et al*. [23] reported the promising results of highly ordered layered GO membranes for the adsorption of Cu^{2+}, Cd^{2+} and Ni^{2+} (62-83 mg/g adsorption capacity). The results also emphasized that the adsorption capacity was greatly affected by the ionic strength. Moreover, Liu *et al*. [24] successfully prepared GO-based membrane crosslinked with polyaluminum chloride (PACl). Experimental results revealed a significant relationship between OH/Al ratio of PACl and the performance of GO-based membranes. Specifically, it can be concluded that the enhanced water flux was associated with the increased of the interlayer spacing as the OH/Al ratio increased. These findings further concluded that both water flux and organic molecules rejection can be achieved by varying the OH/Al ratio.

Significant efforts have also been directed towards exploring the potential of reduced GO (rGO) membranes to overcome issue related to stability and leaching out of GO from membrane matrix [25, 26, 27]. rGO with desired interlayer spacing poses hold immense potential for selectively blocking specific ions and enhancing selective permeability, thereby facilitating superior pollutants removal capabilities. GO with fewer and controllable number of oxygen groups has gained attention for various applications. It is well accepted that the hydroxyl and carboxyl groups were responsible for the formation of uniform rGO membranes where the transport of water and ions permeation mainly dependent on the reduction degree of GO [28]. Similarly, Study from Fan *et al.* [25] have conclusively shown that controlling the hydrothermal reduction time is crucial in regulating the reduction degree of GO, which in turn plays a critical role in improving the permeability and interlayer structure of the resulting membranes. The authors also reported remarkable rejection of Methyl blue, Congo red Crystal violet dyes up to 99%. Recent studies have also highlighted that the inner layer of the self-supported reduced GO can facilitate the transport of Cu^{2+} through the layers, resulting in exceptional adsorption capacity (149 mg/g) [9].

Besides, Chen *et al*. [29] successfully prepared a novel *β*-Cyclodextrin (*β*-CD)-modified GO membranes for removal of Bisphenol-A. The resulting membranes exhibited a tremendous increment in water flux and adsorption of Bisphenol-A. The water flux was also observed to increase linearly with the increase in pressure, indicating the stability of the (β-CD)-modified GO. While rGO has shown a great potential in variety of applications, it's surprising to note that there is still limited number of studies specifically focused on rGO UF membranes. This is a research gap that needs to be addressed to fully understand the potential of rGO-based membranes for wide ranges of applications.

3.0 GO/POLYMER-BASED MEMBRANES

A considerable number of studies have also focused on the modification of UF membranes by the incorporation of GO into polymer-based membranes to enhance the mechanical strength, thermal stability, hydrophilicity as well as the performance of membranes. In the past few years, number of studies have highlighted the advantages of incorporating GO or modified GO into membrane UF matrix [30, 31, 18, 32].

Generally, it is widely believed that the incorporation of GO into the membrane matrix can significantly enhance the membrane hydrophilicity and porosity, thus improving overall membrane performance and efficiency [16, 17, 33, 34, 35]. For example, promising results were reported by Rafal Sitko *et al*. [34] on the effective adsorption of heavy metals ions (cobalt, nickel, copper, zinc, cadmium and lead) by GO/cellulose membranes. The authors emphasized that the chemical adsorption, resulting from the complexation of metal ions with the oxygen containing-groups on the surface of GO plays a crucial role in the monolayer coverage of membranes, ultimately affecting the adsorption capacity. Highest adsorption capacity was revealed for Pb(II) with up to 107.9 mg/g. Zimbianci *et al*. [32] on the other hand, indicated that the performance of polysulfone (PSf) hollow fiber membranes embedded with GO surpassed the conventional granular activated carbon in terms of adsorption capacity for Ofloxacin, Benzophenone-3, rhodamine, dichlofenac and triton x-100. Additionally, the study also evidenced that the loading of GO significantly influenced the performance of the prepared membranes. Similarly, studies by Khumalo *et al*. [36] revealed the superior capability of PSf membranes incorporated with GO for Pb(II) (>80% adsorption). The study also suggested that the adsorption capacity was greatly influenced by the pH of solution. Significant researches also highlighted the leaching issues of GO from membrane matrix due to their high affinity with water [37]. Poolachira and Velmurugan [38] demonstrated that incorporation of polyvinylpyrrolidone (PVP) together with GO into PSf membrane matrix could simultaneously impart better membrane permeability (150 L/m^2 hr) and high removal of Pb^{2+} (80%). The addition of PVP also enhanced the distribution of GO within the membrane surface.

It was reported that the presence of functional groups of the embedded GO is crucial for dyes removal. It was reported that the functional groups of GO membranes could create strong zeta potential which could favourably enhance the repulsive force between dyes and the negative charges of membrane surface, leading to excellent removal of pollutants [17]. However, for polymer-based membrane embedded with GO, the removal of dyes depends on the membrane surface charge, ionic strength and capacity which is promoted by the repulsive force between dyes and the functional groups of GO. Kadhim *et al*. [31] reported the superior performance of PES membranes incorporated with GO for removal of acid black (AB) and rose bengal dyes (up to 99% rejection). Sunil *et al*. [33] incorporated GO into PSf membrane in which the resulting membranes exhibited 100% removal of methyl orange, methyl blue, sulfon black F, Rhodamine 6G and malachite green dyes.

Considerable efforts have also been devoted to the utilization of modified GO for the preparation of adsorptive membranes, aimed at improving their effectiveness in the adsorption of various contaminants from water and wastewater. A study by Wang *et al*., [39] indicated that graphene-oxide polydopamine crosslinker endowed the polyethersulfone-sulfonated polyethersulfone (PES-SPES) membranes with an excellent adsorption properties and good reusability [39]. Moreover, Ismail *et al*. [40] investigated the performance of hybrid UF membranes prepared from PES and quaternized polydopamine anchored reduced GO (QSiPD-rGO) for water treatment. The results showed that incorporation of QSiPD-rGO was beneficial in improving the water flux (up to 270 L/m^2.hr), rejection of BSA (98%) as well as anti-fouling properties. The membranes also demonstrated excellent anti-bacterial activities (against *E. Coli*). Advance technique such as laser reduction was also utilized to produce electrically conductive membrane [41, 42, 43]. Straub *et al*. [43] indicated that the laser-reduced GO membranes is a highly promising technique for designing a tunable and conductive membranes with enhanced performance.

Recently, the incorporation of metal organic frameworks into membrane matrix has also been introduced. Ma *et al*. [44] demonstrated that the incorporation of a modified GO which was successfully anchored with UiO-66, an archetypal metal-organic framework into PES membranes could overcome the layer stacking of GO and impart better permeability and superior anti-fouling performance.

Previous studies have also been directed towards the development of adsorptive membrane with enhanced anti-fouling properties [45]. The addition of GO in UF membrane matrix could enhance the anti-fouling properties of the prepared membranes [31, 46, 47]. In addition, another study indicated that incorporation of ferrihydrite (FH) into graphene oxide (GO) promoted the excellent anti-fouling properties by reducing 95% of the total fouling resistance during the filtration of natural organic matter (NOM). In addition, the prepared membranes also demonstrated an outstanding performance for the removal of carbonaceous DBPs [48]. Yuan *et al*. [10] revealed that nano-GO was able to enhance anti-fouling properties of PSf membranes (with 74% flux recovery ratio). However, the study does not include the adsorption performance of the prepared membranes. An innovative approach was also dedicated for addressing biofouling by incorporating rGO flakes into bacterial nanocellulose (BNC). The resulted membranes exhibited enhanced water flux and anti-biofouling properties as well as remarkable mechanical and chemical stability under a wide range of pH conditions and vigorous mechanical agitation [49]. This anti-fouling approach represents a significant step towards the development of cost-effective and high performance UF membranes [49, 50].

4.0 CONCLUSION

In summary, various efforts have been devoted to the development of highly efficient adsorptive membranes through the utilization of GO/modified GO whether as membrane-based or additive. The primary goal is to enhance the adsorption performance of UF membranes for different types of pollutants including heavy metals and dyes, while also improving the stability and anti-fouling properties of the prepared membranes. Undeniably, the interaction between the functional groups of GO or modified GO and pollutants has a significant impact on the overall adsorption performance. Furthermore, the use of advanced materials such as MOF can be regarded as the recent trend that offer excellent potential for developing membranes with superior removal and adsorption of pollutants and anti-fouling performance. It is anticipated that adsorptive UF membranes would make a remarkable progress towards a viable and innovative route for the removal of various pollutants from water and wastewater.

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