J. Applied Membrane Science & Technology, Vol. 25, No. 1, April 2021, 11–19

Grand Challenges in Membrane Biofouling Mitigation

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Submitted: 21/11/2020. Revised edition: 11/1/2021. Accepted: 11/1/2021. Available online: 25/3/2021

ABSTRACT

Biofouling prevention is a critical challenge in the membrane-based water and wastewater treatment process. Carbon-based nanomaterials which are having exceptional physical, chemical, and electrical properties bring new technologies for addressing biofouling. This paper highlights the novel membrane developments using carbon-based nanomaterials and their potential applications for anti-biofouling technology and its recent cutting-edge developments. Finally, this review also outlines future opportunities for carbon-nanomaterial application in environmental systems.

Keywords: Biofouling, membrane, water treatment, Carbon nanomaterials, electric potential

1.0 INTRODUCTION

Presently, the global water crisis and especially the current water challenges due to accelerating population growth and rapid developments in industrial and agricultural activities worldwide have incited a drastic increase of contaminants that are released into the environment [1]. Membrane technology researchers believe that breakthrough solutions can be achieved through cutting-edge chemical engineering and technology. The most common technology for drinking water production is desalination processes. Membrane technology is now highly used for desalination and water treatment, but the energy and operational costs are high, and membranes do not always remove emerging contaminants. Critical challenges in membrane separations and filtration systems are fouling such as scaling, colloidal, organic, and biofouling, which leads eventually to high energy demand and less economic [2, 3].

Especially, biofouling control is one of the greatest challenges in a pressure driven membrane filtration system in water and wastewater treatments [4]. For instance, only a small number of bacteria that infiltrate the membrane system are sufficient for biofilm development. Biofouling negatively affects the water flux and leads to high energy demand, so to minimize energy consumption, the development of cutting-edge anti-biofouling technology for desalination and wastewater treatment is needed. Although the chemical cleaning (hypochlorite, acid, and base) processes are in practice currently [5], these chemicals most likely to deteriorate the active layer of the reverse osmosis membrane, and thus

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DOI: https://doi.org/10.11113/amst.v25n1.203

selectivity decreases eventually life span decreases and frequent replacements are needed [6].

The coupled nature of many important processes, e.g., the energy required to treat water and the water required to generate energy, makes identifying solutions at the water/energy nexus particularly important [7]. Currently the role nanomaterials and nanotechnology are very important in many environmental applications. Many varieties of nanomaterials having huge potential due to their unique properties such as high surface area and catalytic properties [8]. Electrically conductive nanomaterials, in particular, hold great utility in many applications, including but not limited to the development of energy-storage devices such as batteries, supercapacitors, fouling-resistant systems for desalination and water treatment, enhanced separation methods, and innovative biosensing or electrocatalytic platforms [9,10]. In this short article, the cutting-edge technologies based on carbon nanomaterials and their application in tackling membrane separation issues and solution will be highlighted.

2.0 BIOFOULING IN PRESSURE DRIVEN MEMBRANE PROCESSES

Microorganisms in wastewater treatment systems tend to adhere to membrane surfaces and form a gel layer called biofilm, which serves as a secondary membrane in the separation process. An initial deposition of planktonic bacteria on solid surfaces, which overcomes the repulsive forces between the bacteria cell and surface, initiates the biofilm formation. It causes an increase of fluid friction resistance on the feed water side, which increases feed pressure ΔP_{feed} and also overall hydraulic resistance of the membrane $\Delta P_{membrane}$ [11]. Treatment to remove fouling includes cleaning in place (CIP) by using strong bases, acids, and oxidizing chemicals (i.e. chlorine) that are specific to the fouling material [12]. Bacterial adhesion and biofouling is a serious problem in water filtration membrane systems, industries, and medicinal equipment [13]. It is very important to minimize the organic carbon from wastewater, thus limiting the major substrate for the growth of biofouling.

3.0 ROLE OF NANOMATERIALS AND NANOTECHNOLOGY

Many antimicrobial nanomaterials are studied including their merits and demerits towards biofouling control and disinfection of microbes in water [14]. However, once the deposition of foulants has taken place, the surface modification is no longer effective to prevent fouling, because the effect of solute-membrane interaction is severely reduced once a layer of deposited foulants is formed [14]. An emerging new membrane field is composite polymeric membranes embedded with nanomaterials. Among all nanoparticles, carbon nanotubes (CNTs) and graphene have a great potential application in water and wastewater treatment. Intrinsic properties of these carbon nanomaterials have high physical properties and chemical resistance to chemicals, combined with electrical conductivity and ductility, and antibacterial activity.

A recently discovered material by Prof. Tour's group named laser-induced graphene (LIG), a facile and scalable approach to produce conformal 3D porous graphene structures on surfaces in a single step [15]. LIG is a desirable material for use in water desalination and membrane technologies for several reasons. While its extensive, multi-layered graphitic structure confers the impermeability characteristic of pristine graphene, the hierarchical LIG structure is also porous, allowing for the possibility of water permeation with concomitant salt rejection; moreover, the effective pore size can be successfully modulated by the choice of polymer substrate for laser scribing, laser irradiation conditions, and the dopants or functional molecules present within the LIG network [9]. Then our group has developed the fabrication method so that various LIG forms can be obtained on almost any carbon substrate, enabling graphene coatings on a wide range of materials [16]. LIG can be chemically tuned or functionalized and has enabled the production of biocides and oxidant and reductant radicals with exceptional electrochemical performance as electrocatalytic surfaces and aided the development of various technologies for water remediation [9]. More recently, polymer films and membranes decorated with standalone or doped LIG have exhibited remarkable antimicrobial and biofilm inhibition activities while conducting current in aqueous environments, paving the way for the development of energy-efficient graphene surfaces and filters suited to many disinfection and water treatment systems [9, 17].

4.0 CUTTING-EDGE ANTIBIOFOULING TECHNOLOGIES

Electrically conductive solid surfaces such as surgical stainless steel [18], gold [19], Indium tin oxide (ITO) [20], platinum [21] were extensively studied for avoiding biofilm and bacterial attachment in the medical and industrial field using low electric potential. An electroconductive feed spacer was also studied as a tool for controlling biofouling in a lab-scale crossflow membrane system at a low electric potential of 1 V [22]. Applying electric potential on membrane modules to control biofouling is environmentally friendly and can be potentially automated and applied to highly electrically conductive membranes. In recent years both alternating (AC) or block current and direct current (DC) have been widely studied, including the specific role of anodic, cathodic and block current to detach, prevent attachment and inactivate the bacterial cells [20,23]. Furthermore, by applying a low alternating electrical potential on the conductive membrane, biofilm growth could be controlled and inactivated [24].

Upon application of cathodic current on CNT self-supporting membranes, bacterial attachment on the membrane surface decreases significantly however, bacteria that remained on the electrode surface were alive. In contrast, when an anodic current was applied, the bacteria that remained on the surface became inactive with time, although bacterial detachment was not significant. But both detachment and inactivation were effectively noticed in the alternating current. Hence, an alternating (block) electric field has been suggested as a possible means of controlling biofilm development [20,25]. The alternating current had a synergic effect of both anodic and cathodic potential.

Many electrically conductive CNT composite membranes are developed for biofouling prevention under an electric field [26,27]. Examples include de Lannoy *et al*., who developed an electrically conductive CNT-polyamide composite membrane with an interfacial polymerization process and prevented the formation of microbial biofilms using 1.5 V alternating current (AC). Ronen *et al.* studied electrically conducting composite CNT membranes for bacteria detachment by applying a direct current (DC) potential of 1.5 V [26]. Our research group recently studied the effect of low voltage electric fields, both AC and direct current (DC), on the prevention of bacterial attachment and cell inactivation to highly electrically conductive (40000 S/m) self-supporting CNT membranes [4]. Enhancing the antifouling effect can be achieved using other strategies as well, for example, CNT membranes anchored with silver ions are effective in reducing biofouling in membrane separation processes [28].

For LIG surfaces, an initial biofouling investigation showed antibiofilm properties but antimicrobial activity was limited to support from an electrical potential [29]. PES-LIG, PPSU-LIG surfaces, and PES-LIG porous membranes exhibited exceptional antibacterial and anti-biofouling properties when tested with *P. aeruginosa* and mixed bacterial culture [30]. The PES-LIG membrane fabricated using a commercial ultrafiltration membrane as a substrate showed complete elimination of bacterial viability in the permeate (6-log reduction) in a flow-through filtration mode under an electric field. The LIG composites also demonstrated similar antibacterial and anti-biofouling activity [17,31]. Similar to CNTs, LIG with embedded silver nanoparticles showed an anti-biofouling effect and greatly increased the surface toxicity toward bacteria [32].

The bacterial adhesion and attachment are mainly based on the interaction forces such as electrostatic repulsion/attraction, van der Waals attraction, and hydrophilic/hydrophobic forces, it depends also based on the bacteria surface properties [33]. Another reports stats that electrostatic and electrophoretic forces are dominating the bacterial attachment when –ve potential applied to the solid surface, while applying +ve potential the bonding between the solid and bacterial surfaces becomes loosening due to osmotic forces, it could be washed off with higher shear forces [34]. Low voltage current from 60-100μA was studied in detail for avoiding the adhesion for gram (-) and gram (+) bacteria in solid surfaces [18,35]. Most importantly, it has to be taken into account that permeates drag forces play a significant role in biofilm adhesion [36].

It is well known that graphene-based materials exert intrinsic antimicrobial activities. For instance, graphene and graphene oxide nanosheets display electrochemical and contact-based toxicity toward both Gram-positive and Gram-negative bacteria, phytopathogens, parasitic algae, and several other classes of microorganisms [37]. Besides, surfaces consisting of graphene derivatives are refractory to biofouling and the formation of microbial biofilms [38]. The microbicidal and anti-biofouling activity of graphene surfaces is enhanced further by the introduction of an electrical current. The inactivation occurs in two ways, direct oxidation, which damages the cell wall of the membrane, and indirect oxidation due to the generation of biocides such as oxide radicals (OH^-) and chlorine (Cl^-) and hydrogen peroxide (H_2O_2), etc. [39]. Biofouling prevention and inactivation mechanism were explicit in detail. H_2O_2 prevents biofilm growth on solid surfaces, however, H_2O_2 is not detected in the bulk solution. It is supposed to be nearer to the solid surface. In a high electric field complete inactivation was observed, this is obviously due to direct oxidation and also the generation of biocides. The bacterial cell damage during the inactivation is elucidated in detail for different treatments including high hydrostatic pressure, pulsed electric fields, and thermos sonicated showed a total disruption of the cell membrane, perforation, and release of the cell wall and formation of pores [40].

LIG electrodes also kill microbes via oxidative stress mechanisms, through direct contact as well as by promoting the evolution of reactive chemical species. For instance, according to Singh *et al.* [29], the cathodic oxidation of water molecules formed considerable amounts of H_2O_2, which is a potent oxidizing agent; by reacting with key biomolecules and redox-state mediators in microorganisms, H_2O_2 and similar chemical species generated electrochemically can engender substantial oxidative stress that leads to cell death. The edge-rich and hierarchical architecture of LIG is especially integral to anti-biofouling effects, as crushed LIG exhibits markedly reduced efficacy in preventing bacterial adhesion and biofilm materialization [41]. Initially adhering bacteria are known to adhere more reversible than bacteria growing in the later stage of biofilm formation, hence prevention of initial attachment is very important [4].

5.0 SUMMARY AND FUTURE PROSPECTS

Carbon-based nanomaterials and their remarkable physical and chemical properties could make next-generation electroconductive biofouling resistant membranes. These carbon-based nanomaterials are not only limited to the separation process, this also can apply to novel electrocatalytic platforms by doping different metals [42, 43], fog harvest, membrane distillation [44–46] resource recovery, and energy storage applications like batteries and supercapacitors. However, making a robust electroconductive membrane with retaining electrical conductivity is still a challenge. Secondly, protection of oxidation during the electrochemical reaction another important challenge which is partially protected with polyaniline coating [47]. The further developments of these carbon-based materials to overcome these obstacles will be beneficial for efficient and attractive low-cost water treatment and other environmental applications.

ACKNOWLEDGEMENT

Dr. Chidambaram Thamaraiselvan is grateful to the Department of Science and Technology (DST), New Delhi, India, for awarding the INSPIRE Faculty Award (DST/INSPIRE/04/2019/000925).

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