Desalination xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Engineering Advance

Recent advances in the development of (bio)fouling resistant thin film composite membranes for desalination☆

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ARTICLE INFO

Available online xxxx

Keywords:
Thin film composite
fouling
surface modification
polyamide

ABSTRACT

Membrane (bio)fouling is a major obstacle to many separation and purification processes. Due to the inherent physicochemical properties of some thin film composite membrane surfaces such as polyamide, these are prone to (bio)fouling. Hence, this review highlights recent advances in the design and development of highly resistant thin film composite membrane through surface modification by either coating or grafting with antifouling polymers and/or antimicrobial polymers/biocidal inorganic nanoparticles.

Contents

1. Introduction

(Bio)fouling, denoted as the "Achilles heel" of membrane process, still remains as one of the most technical challenges in the desalination industry, resulting in a decrease of permeate flux, shortens the membrane lifespan and subsequently increases the operational cost [1,2]. Membrane (bio)fouling is a dynamic process of microbial colonization and growth at the membrane surface [3,4]. Once permanently attached, these organisms start to produce extracellular polymeric secretions (EPS) comprising proteins, glycoproteins, lipoproteins, polysaccharides and other biomacromolecules [5]. The accumulation of EPS and reproduction of bacteria would lead to the formation of mature biofilm [6–8].

In particular, thin film composite (TFC) membranes have currently been used as the primary choice to desalinate seawater. They generally consist of an ultra-thin polyamide (PA) layer which is interfacially polymerized onto a microporous substrate [2,9]. The inherent membrane surface characteristics of the PA TFC membrane such as surface

☆ This is an Engineering Advance paper

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http://dx.doi.org/10.1016/j.desal.2015.06.001

roughness, hydrophilicity and surface charge are, therefore, reported to be strongly related to foulant adhesion. It is generally well-acknowledged that foulants can adsorb to the membrane surface by hydrophobic interaction, van der Waals attraction, hydrogen bonding, Lewis acid-base interaction and electrostatic repulsion [10,11]. Owing to the aforementioned issues, prevention or reduction of undesired interactions between foulants and the membrane surface can be an imperative method to control the adhesion of foulants. This could be achieved through surface modification of commercial TFC PA membrane with the aim to increase the PA surface hydrophilicity, reduce membrane surface roughness and to modify membrane surface charge that has the same electrical charge as the foulants [4]. Hence, this review highlights recent advances in the development of TFC membranes through surface modification. To date, several routes have been proposed to improve fouling resistance including, physical adsorption or coating over the membrane surfaces, covalent attachment of anti (bio)-fouling polymers, and incorporation of biocidal inorganic particles on the membrane surface.

2. Surface modification strategies for TFC membranes

2.1. Anti-adhesion approaches

2.1.1. Polyethylene GLYCOL (PEG)ylated polymers

Imparting PEGylated materials has been commonly employed due to their superlow-fouling ability to combat the nonspecific protein adsorption and cell adhesion [12–14]. PEG is highly hydrophilic and neutrally charged. It is also able to form hydrogen bonds with water that could increase surface hydrophilicity and lower the interaction with nonspecific foulants [12,15].

In recent advances, novel low fouling TFC membranes were successfully fabricated by in situ PEGylation of polyamide composite membranes [15,16]. Resembling the *m*-phenylenediamine (MPD) structure, commonly used monomer to prepare polyamide RO membrane, amine terminated hydrophilic PEG (MPD — PEG — MPD or MeO — PEG — MPD) monomers as shown in Fig. 1 were used to react with trimesoyl chloride (TMC) during the composite membrane fabrication [16]. Similarly, in situ PEGylation of poly(piperazineamide) TFC nanofiltration membranes was performed by interfacial polymerization between TMC and poly(piperazine) (PIP) + PIP — terminated PEG (PIP — PEG — PIP) and PIP + MPD-terminated PEG (MPD — PEG — MPD) and PIP + alkyl amine terminated — PEG (H_2N — PEG — H_2N) mixtures (as can be seen in Fig. 1), respectively [15]. The PEGylated TFC membranes exhibited superior antifouling property owing to the hydrophilization of polyamide network by PEG, reduced surface roughness and probably due to effect of steric hindrance by covalently attached PEG chains than that of non-PEGylated TFC membranes [15,16].

However, some problems still need to be addressed. Although PEGylated materials demonstrated excellent protein resistance ability, PEG could decompose in the presence of oxygen and transition metal ions found in most biochemically relevant solutions [13,17,18].

2.1.2. Natural hydrophilic polymer sericin

Natural hydrophilic polymer sericin, as shown in Fig. 2, is a water-soluble globular protein having polar side groups of hydroxyl, carboxyl and amino groups [19,20]. The polymer sericin was coated on the surface of commercial TFC RO membranes followed by cross-linking with glutaraldehyde (GA) [19]. The siricin-coated membrane showed decreased water permeability due to the additional hydraulic resistance, but improved salt rejection as a result of the enhancement of surface negative charge. Resistance to BSA fouling was improved due to enhanced surface hydrophilicity, increased surface negative charge and smoothed surface morphology [19]. Besides, Zhou et al. [20] used the natural hydrophilic polymer sericin to react with TMC during the interfacial polymerization process. The fouling experiment demonstrated that the sericin-TMC composite membrane possessed better fouling resistances to both BSA and sodium alginate (SA) when compared to the commercial composite nanofiltration membrane (NF270). This is mainly due to the higher electrostatic repulsion between the foulant molecules and negatively charged sericin-TMC membrane that resulted in less adsorption of foulant molecules on the membrane surface [20].

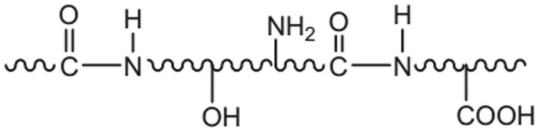

Fig. 2. Schematic diagram of natural polymer sericin chemical structure [19,20].

2.1.3. Hyperbranched polymers

Surface coating with polymers possessing hydrophilic end groups, dendritic or hyperbranched polymers, is of great interest to researchers in order to impart protein resistance to the surface of TFC membrane [21,22]. A large number of functional groups and low solution viscosity cause the hyperbranched polymers to be advantageous for various applications [22]. Nikolaeva et al. [22] used hydrophilic hyperbranched poly(amido amine) (PAMAM) for the surface modification as represented in Fig. 3. Highly reactive acid chloride groups are, therefore, used for the covalent bonding of PAMAM to the PA layer by the formation of amide linkages between TMC moieties of the PA layer and amine groups of PAMAM molecules. The modification is accomplished by spraying a 10 wt.% solution of PAMAM onto the PA surface of using either methanol (PAMAM1) or water (PAMAM2) as solvent. In comparison to the unmodified membranes, both modifications led to a significant increase in water flux which is attributed to the suppression of subsequent cross-linking during the final curing step. In view of salt rejection and protein adsorption, the use of water (PAMAM2) has been found to be beneficial over the use of methanol (PAMAM1). This is primarily due to the formation of an additional highly hydrophilic PAMAM layer, which can be recognized as a hydrogel layer when in contact with water. Additionally, PAMAM can be synthesized in a simple one-pot polymerization and is also easily purified, making it a low cost material [22].

2.1.4. Zwitterionic polymers

Zwitterionic polymers have also drawn great attention as a new generation of fouling resistant material in recent years [23–25]. They comprise both positive and negative charged units that can create stronger and more stable electrostatic bonds with water than hydrophilic materials [23]. Inspired from the adhesive proteins found in mussel, a multifunctional zwitterionic amino acid L-DOPA (3-(3,4-Dihydroxyphenyl)-L-alanine) has been successfully anchored on the

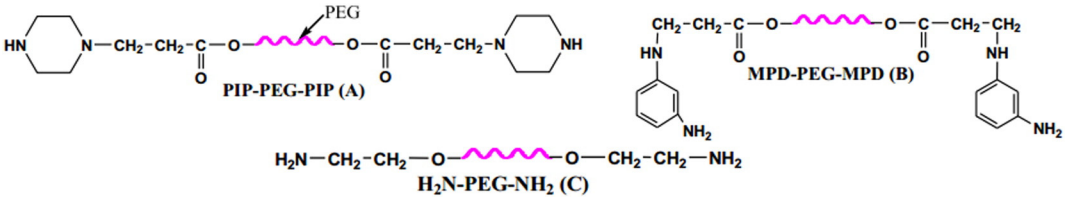

Fig. 1. Amine terminated hydrophilic PEG monomers [15,16].