



Surface modification of polyamide RO membrane for improved fouling resistance



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HIGHLIGHTS

- The degree of fouling was indicated by the values of flux decline index (m) and MFI.
- The fouling of GPPTMS-modified RO membranes would be reduced.
- The fouling resistance was increased by using a hydrophilic epoxy compound.

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ABSTRACT

This study investigates how surface modification of RO membranes enhances resistance to fouling. The standard mechanisms of membrane fouling are extended to identify RO pretreatment processes. The degree of fouling resistance of modified RO membranes are studied with respect to the modified fouling index (MFI). According to the definition of the MFI, the higher the MFI value of an RO membrane indicate the greater the fouling resistance. A polysiloxane system consisting of 3-glycidoxypropyltrimethoxysilane (GPPTMS) modified on a PA surface displays a complex structure. The fouling resistance of the GPPTMS-modified RO membrane was increased by using a hydrophilic epoxy compound, which forms hydrolyzed functional groups. The flux decline index (m) and the MFI decreased for GPPTMS concentrations less than 1.0 wt%. These results prove that fouling of GPPTMS-modified RO membranes would be reduced when the concentration of GPPTMS was over 1.5 wt%. The values of m and MFI for RO membrane are much higher than those for GPPTMS-modified RO membranes because more foulants are brought to the membrane surface.

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1. Introduction

Reverse osmosis (RO) technology is increasingly used for desalination, waste water treatment, water reuse and so on. It is increasing globally due to continuous process improvements, which in turn lead to significant cost reductions. The enhanced mechanical, biological and chemical strength of the aromatic polyamide RO membranes, as well as the increased permeability, have reduced the membrane cost per unit volume [1,2]. However, RO membranes are weak for bio-fouling due to hydrophobic and rough surfaces. Bio-fouling can cause obvious increases in hydraulic resistances and substantial decreases in membrane permeate fluxes. Membranes with hydrophobic surfaces are susceptible to organic fouling [3–5].

Improving membrane anti-biofouling properties are the main challenges for the aromatic polyamide RO membranes to realize high performance over a long period of operation. Surface modifications such as physical adsorption and chemical bond formation are potential methods to improve membrane anti-biofouling properties. It has been reported that the chlorine resistances of the aromatic polyamide RO membranes can be improved by coating with high chlorine-resistance polymers (such as poly(*N,N*-dimethylaminoethyl methacrylate) [6], poly(vinyl alcohol) [7,8] and hexafluoroalcohol-containing polyamide layer (HFAPA) [9,10] which cover the active sites of aromatic polyamides and protect them from chlorine scouring. An anti-biofouling aromatic polyamide RO membrane with photocatalytic destructive capability on microorganisms was designed by self-assembly of TiO₂ nanoparticles onto membrane surface [11,12].

Belfer et al. [12,13] have approached the problem of membrane fouling by developing an in-situ method for surface modification of commercial RO membranes. In this method, redox-initiated grafting of acrylic or vinyl monomers is used to modify the surface of the membrane with polymers containing various functional groups. In the

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current study we set out to determine how modification of membrane surfaces by means of the above described method affects antifouling and performance properties of a range of RO membranes, from seawater (SW), through brackish water (BW), to ultra-low-pressure (ULP) membranes.

Foulants can be classified into four categories: adsorbed organic compounds, biological growth, metal (hydr)oxides, and particulate matter. Particulate foulants can be further classified into suspended matter, microorganisms, and colloids. These foulants can be reduced by pretreatment of the feed water [14]. A biofilm caused by biofouling is difficult to remove because it protects microorganisms against the action of shear and disinfecting chemicals. In addition, remaining biofilm leads to rapid regrowth [15]. Membrane fouling can be reduced by appropriate pretreatment of the feed stream and/or chemical modification of membrane surfaces. Active layers of the modified membranes can reduce the adsorption of organic matter, thus preventing subsequent biofouling.

In a previous work, we introduced a method to improve salt rejection by treating polyamide TFC RO membrane through sol-gel reaction [16,17]. Treatment of polyamide TFC RO membranes with OctES (octyltriethoxysilane) significantly increased salt rejection. This paper focuses on the relationship between the structure of silane coupling agents and the properties of RO membranes. We selected silane coupling agent 3-glycidoxypropyltrimethoxysilane (GPPTMS) which is one of the common precursors for the preparation of hybrid organic-inorganic polymer. GPPTMS is also commonly used as a grafting agent to surface modification [18]. The chemistry of GPPTMS has a hydrophilic epoxy group and flexible carbon chain. It can be expected more hydrophilic property as surface modification on membrane surface. Athanasekou et al. [19] has reported that GPPTMS can be applied for wastewater treatment.

This study investigated how the surface modification of RO membranes enhances resistances to fouling. The standard mechanisms of membrane fouling are extended to identify pretreatment processes for RO membranes. Also we study the degrees of fouling of modified RO membranes, and these are assessed using fouling decline index (m) and modified fouling index (MFI) values, respectively.

2. Theory

2.1. Flux decline index

The RO flux decline can be represented by the following empirical relation [20]:

$$F_t = Kt^m \quad (1)$$

where F_t is the flux at time t , K is a constant, and m is the log-log flux decline index. Eq. (1) predicts that a log-log plot of flux vs. time will yield a straight line, with a negative slope of value m . The constant K is generally taken to be the initial flux value, F_0 , thus the flux decline index can be expressed as:

$$m = \log(F_t/F_0)/\log(t/t_0) \quad (2)$$

where t_0 is the initial measurement time. It means for measuring the time of initial performance for the membranes.

2.2. Modified fouling index for RO membrane

The MFI, which is derived from the silting index and the silt density index, is based on fouling in gel filtration [21]. These parameters are often applied to RO membranes to measure the fouling potential, and they are also used in microfiltration and ultrafiltration applications. The disadvantage of these fouling tests is that they are based on dead-end filtration, but they are applied in a cross-flow mode. This means

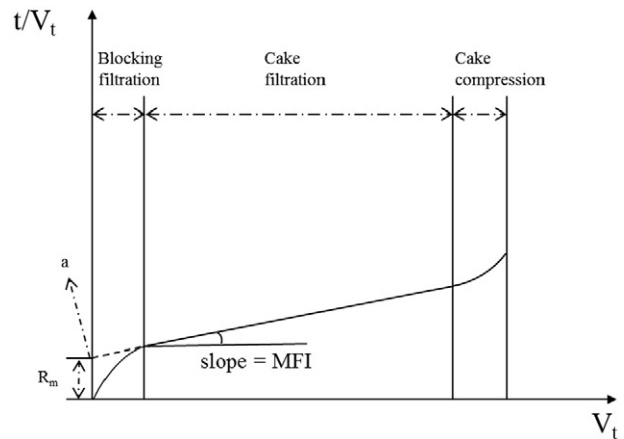


Fig. 1. Ratio of filtration time and filtrate volume as a function of total filtrate volume [20].

that the flow conditions in the module are not taken into account, but this is a crucial parameter in process optimization. The MFI can be represented as shown in Fig. 1, providing the thickness of the cake layer formed on the membrane surface (fouling) is proportional to the filtered amount. To determine the MFI value, the flow rate (Q) is obtained by multiplying the measured permeation flux by the membrane area (A). The MFI is based on the fouling resistance, which is assumed to be proportional to the filtered volume V after filtration time t :

$$\frac{dV}{Adt} = \frac{\Delta P}{\eta(R_m + R_c)} \quad (3)$$

After integration,

$$I \frac{t}{V} = \frac{\eta R_m}{A \Delta P} + \frac{\eta I}{2A^2 \Delta P} V \quad (4)$$

The resistance of the cake (R_c) is assumed to be independent of the applied pressure. V/A is the permeate volume produced per membrane area (A), and I is a proportionality factor:

$$R_c = \frac{V}{A} \times I \quad (5)$$

The MFI is calculated as

$$a = \frac{1}{J_0} = \frac{\eta R_m}{\Delta P} \quad (6)$$

and

$$\frac{1}{Q} = a + MFI \times V \quad (7)$$

where $J_0 = \Delta P/\eta R_m$ is the initial flux at time $t = 0$ and ΔP is the applied pressure. The resulting value is multiplied by the filtration time (t) to obtain the filtered amount (V). By assigning the filtered amount (V) to the horizontal axis (x) and $1/Q$ to the vertical axis (y), the slope of the t/V versus V curve is the MFI value [11,22–23]. A plot of t/V as a function

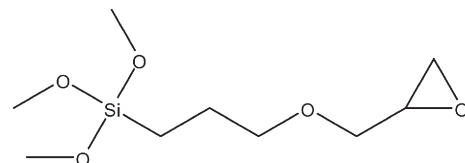


Fig. 2. Molecular structure of 3-glycidoxypropyltrimethoxysilane (GPPTMS).

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