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Improvement of performance and antifouling properties of reverse osmosis membranes using green additive

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ABSTRACT

Aqueous extract of sunflower (*Helianthus annuus*) seeds was used as a novel environmental friendly additive to improve the performance and anti-biofouling properties of cellulose acetate reverse osmosis (RO) membranes. Modified membranes were characterized by Fourier transform infrared spectroscopy and scanning electron microscopy, while the anti-biofouling properties were studied by water contact-angle measurements, water content measurements, and static protein adsorption. The effect of modification of the membrane on the salt rejection and water flux was studied using a cross-flow RO unit. The results indicated that modified membranes have lower contact angle accomplished with high water content. They showed limited adsorption of protein and microbes, in addition to increased salt rejection and water flux values.

Keywords: Cellulose acetate membrane; Natural extract; Biofouling; Protein adsorption

1. Introduction

Water scarcity continues to remain a big challenge faced by mankind in many regions around the world [1]. Many decades of successful implementations demonstrate how desalination technology can provide supplementary or main water sources. Technological advances in reverse osmosis (RO) membranes during the past decade have significantly reduced the energy cost of water production via desalination [2].

However, biofouling resulting from the attachment of microorganism communities to the membrane surface is the major obstacle for the widespread application of membrane technology [3]. The biofouling adds filtration resistance and increases the operational costs because of the need for frequent cleaning and maintenance [4,5]. The use of biocides and cleaning protocols for biofouling control may be reduced by membranes resistant to biofouling. Hydrophilic and smooth surfaces have historically shown resistance to protein and bacterial adhesion. Cellulose materials are located on the high list of polymers used for preparing organic membranes. Cellulose acetate (CA) is considered as the most common membrane materials used for water application due to its natural characteristics, low cost, extraordinary potential flux [6–9], long life time, less clean requirement, and minimal fouling membrane via its high hydrophilicity [10,11]. Recently, polydopamine has emerged as a hydrophilic surface modification agent capable of reducing oily water membrane fouling [10,12].

The use of chemical compounds in a variety of applications is conditioned by environmental and health

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concerns. Accordingly, industrial requirements for chemical compounds refer not only to their efficacy but also to their safety [13,14]. Plant extracts are viewed as a rich source of natural chemical compounds that can be extracted by simple procedures with low cost. During the last years, plant extracts were employed as scale inhibitors in order to develop new cleaning chemicals for green environment [15–17]. The data indicated that these extracts act as good antiscalants. The efficiency of these extracts was attributed to the formation of an inactive complex of calcium with the constituents of the extracts. This complex poisons crystal nuclei and inhibits the growth of the scale [16]. Also, several plant extracts have been tested as inhibitors for the corrosion of some metals in different media using chemical and electrochemical techniques [18–25]. The mechanism of the inhibition of the corrosion of metals with the plant extracts was attributed to (a) adsorption of the chemical constituents of the extracts at the metal/solution interface and (b) the formation of metal complexes at the metal/solution interface [18]. Furthermore, plant extracts were employed to improve the corrosion resistance of the organic coat on the steel surface [26–28].

The main objective of this work is to improve the performance and antifouling properties of CA-RO membrane by adding extract of sunflower (*Helianthus_annuus*) seeds during membrane preparation process using phase-inversion technique. The modified membranes will be characterized by Fourier transform infrared (FTIR), contact angle, and scanning electron microscopy (SEM) techniques. The performance of the pristine and modified membranes will be assessed for water desalination using water flux and salt rejection measurements.

2. Materials and methods

2.1. Materials

CA (molecular weight of 100,000 g mol^{-1} and 39.8 wt.% acetyl) was received from Aldrich, USA and 1,4 dioxane was supplied by Panreac Quimica S.A (Barcelona, Spain). Methanol (purity = 99.5%) and acetone (purity = 99%) were received from Labsolve (Lisbon, Portugal). Acetic acid (purity = 99.8%) was supplied by BDH Anala R (England), and NaCl was purchased from Merck (USA), Aldrich (USA), and the Egyptian Petrochemical Company, Egypt. Egg Albumin powder (purity = 99%) was supplied from Raheja Center, Mumbai.

2.2. Preparation of extracts

Stock solution of *Helianthus annuus* extract was obtained by drying the seeds for 2 h in oven at 70°C and grinding to powdery form. A 5 g sample of the powder was refluxed in 100 mL distilled water for 1 h. The refluxed solution was filtered to remove any contamination. The stock solution was evaporated to obtain extract residue.

2.3. Preparation of CA-RO membranes

CA-RO membranes were prepared using the phase-inversion method. Mixture of dioxane (27 g), acetone (13.5 g), and acetic acid (5 g) were used as solvents for CA (8.45 g), while methanol (10.7 g) was used as a nonsolvent. Different concentrations (0.03, 0.3, and 0.6 g) of *Helianthus annuus* extract were added to this mixture to prepare modified CA-RO membranes. This mixture of solution was left under stirring for 24 h at room temperature, until CA completely dissolved. The solution was put into an ultrasonic bath for 30 min to remove the air bubbles entrapped in the polymer solution. The RO membranes were obtained through the spreading of the solution onto a glass plate using a knife of an automatic applicator (Zehntner 2300-Swiss, Switzerland), at room temperature of 42% relative humidity. The thickness of the film was previously selected (250 μm) and spread at a constant speed (10 mm s^{-1}). After casting the solution with evaporation time of the solvent for 60 s, the CA membrane was immersed for 15 min in a deionized water ice bath. The formed membrane was then placed in a water bath at about 4°C for 2 h to eliminate the effect of capillary pressure and then washed with distilled water to completely remove the residual solvents [29]. The prepared CA-RO was then post-treated for 10 min at about 80°C–85°C, then soaked in deionized water for 24 h, and air-dried for 24 h before characterization. Each membrane was prepared three times for reproducibility.

2.4. Characterization techniques

2.4.1. Chemical structure

FTIR spectrometer (FTIR LX 18-5255 Perkin Elmer, USA) was used for characterizing the membranes. The spectra were recorded in the wave number range of 4,000–400 cm^{-1}. The CA membrane samples were ground with KBr in a ratio of 1:10 in the powder form to reduce the particle size; otherwise, the large particles will scatter the infrared beam and cause a slope in the baseline of the spectrum [30]. The powdered mixture was then pressed in a hydraulic press to form a translucent disk through which the beam of the spectrometer can pass.

2.4.2. Morphological investigation

Morphology of membranes was observed using SEM (XL 30 JEOL, Japan). Membranes were fractured in liquid nitrogen for cross-sectional images. The membranes were sputter coated with a thin film of gold under vacuum prior to morphological examination.

2.4.3. Hydrophilicity measurements

2.4.3.1. The contact angle

The contact angle of the CA-RO membrane surfaces was measured using Rame hart, Instrument, France. A drop of distilled water (2 μL) was placed on the membrane surface (3 cm× 2 cm) using a microsyringe (Hamilton Company, Reno, NV, USA). The contact angle was measured within 10 s after the water drop was placed, and the contact angles were measured at five different positions.

2.4.3.2. Water content

Water content is the ratio, expressed as a percentage, of the mass of "pore" or "free" water in a given mass of

the membrane to the mass of the dry membrane [1]. Water content of the membranes was obtained after soaking membranes in water for 24 h, and the membranes were weighed followed by mopping them with blotting paper. The wet membranes were placed in an oven at 85°C for 24 h, after that they were placed in oven under vacuum, and the dry weights of the membranes were determined. The percentage of water content (WC%) was calculated using the Eq. (1):

$$\mathrm{WC\%} = \left(W_w - W_d\right) \times \frac{100}{W_w} \tag{1}$$

where W_w and W_d represent the weights of wet and dry samples, respectively.

2.4.4. Membrane performance

The performance tests (salt rejection and water flux) were conducted for membrane sample (area 42 cm^2) using a crossflow RO unit (CF042, Sterling, USA) equipped with hydra-pump, pressure control valve, and gauge through rejection line, with variable frequency drive (SV015IG5A-4) and flow meter (F-550 USA). Saline salt solutions of NaCl of 10,000 ppm and pH 7 were used. The determination of the total dissolved salt of the permeate water was measured with a pH and conductivity meter (430 portable, Jenway, England). The water flux (F) and salt rejection (R) values were obtained using Eqs. (2) and (3) [31]:

$$F = \frac{V}{A} \times t \tag{2}$$

where V is the total volume of water passing through the membrane (L), A is the membrane area (m^2), and t is the time (h).

$$R = \left(C_0 - C_{\mathrm{memb}}\right) \times \frac{100}{C_0} \tag{3}$$

where C_0 is salt concentration in the feed water side and C_{memb} is the salt concentration in the permeate side of the membrane.

2.5. Antifouling property measurements

2.5.1. Static protein adsorption of the membrane

Adsorptive fouling was evaluated by immersing the membrane in protein feed solution, and the amount of adsorbed protein was determined by measuring the concentration of the protein solution before and after adsorption [3].

In this test, egg albumin powder (1 g) was dissolved in 10 mL phosphate buffer solution (pH = 7.1). To test the protein adsorption, membranes were cut into pieces 2.5 cm× 2.5 cm in size, and 2 pieces of the samples were put into a glass vial containing 10 mL of the protein solution. The vials were vibrated in a shaking bath at a constant temperature of 25°C for 24 h to reach the protein adsorption equilibrium. The concentration of protein in the solution before and after adsorption was measured using UV spectrophotometer (UV-1601, Shimadzu, Japan). The relative protein adsorbed (RP%) was used to identify the extent of adsorptive fouling.

$$\mathrm{RP\%} = \frac{\left(C_0 - C_a\right)}{C_a} \times 100 \tag{4}$$

where C_0 and C_a are protein concentrations before and after membrane soaking, respectively.

2.5.2. Fouling attachment monitoring

For fouling investigation, different membrane samples, 2.5 cm× 2.5 cm in size, were submerged in Mediterranean seawater for 15 d. The dried membrane surface was observed by SEM after coating with a gold layer. The density of the microbes on the membrane surface was investigated from the SEM images.

3. Results and discussion

3.1. Structure investigation of CA-RO membranes

FTIR has been used as a useful tool in determining specific functional groups or chemical bonds that exist in a material. Fig. 1 illustrates FTIR spectra of pristine and modified CA membrane. The band at 2,940 cm^{-1} represents the aliphatic C–H group, and the band at 1,748 cm^{-1} is attributed to the stretching vibration of C=O bond, while the band at 1,636 cm^{-1} represents free water molecule (water of crystallization). The characteristic bands around 1,434 cm^{-1} represent the bending vibration of CH_2, and a band at 1,372 cm^{-1} shows C–H bending vibration. Bands at 1,234 and 1,054 cm^{-1} are assigned to stretching vibration of the C–O bond and C–O–C ether linkage from the glycoside unit, respectively, while 908 cm^{-1} band is corresponding to the out-of-plane C–H bend (pyranose ring) [32–34].

For membrane containing *Helianthus annuus* extract, broad band at 3,374 cm^{-1} was observed. This band can be attributed to the stretching of the hydroxyl group and N–H broad band. The appearance of N–H band's vibration at 3,300–3,500 cm^{-1} is an indication of the presence of amine group [1] as a result of

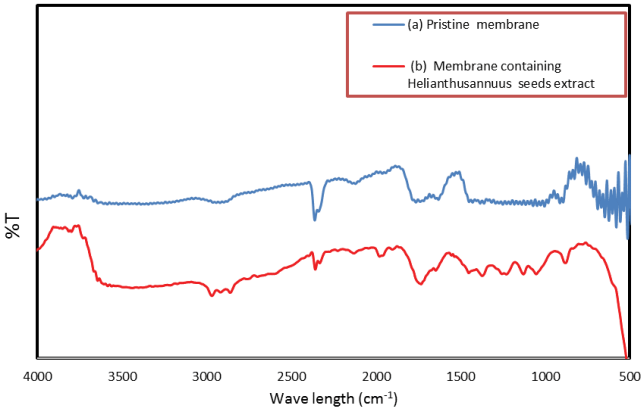

Fig. 1. FTIR spectra of cellulose acetate membranes in the absence and presence of *Helianthus annuus* seed extract.

the presence of amino acids in *Helianthus annuus* extract [35]. Also, the peak at 1,273 cm^{-1} shows N–C bond stretching [1]. The band at 1,651 cm^{-1} is probably attributed to H–O–H bending group of the absorbed water [36], and the band at 1,731 cm^{-1} is corresponding to the C=O bond of the carboxylic group. In addition, the peaks at 2,932 cm^{-1} is assigned to the C–H bending, and the band at 1,049 cm^{-1} probably corresponds to the stretching modes of C–O ether. It is reported that studied extract is rich with flavonoids, triterpenoids, and oils which contain carboxyl and amine groups in addition to phenolic structures [35,37].

3.2. Membrane morphology

The membrane surface and cross-sectional morphologies of prepared membranes were examined using SEM in Fig. 2. As seen, the surface of pristine membrane shows nonporous structure which is typical to CA membrane and has been reported in literature [1] and consisted of a skin layer on the top surface and finger-like pores under this layer. The finger-like macrovoids were visible in the middle of the cross sections. These finger-like structures appeared to be aligned along the direction of solvent/nonsolvent interface, indicating that the phase separation was induced by the nonsolvent penetration of the top surface of the membrane [3]. In the finger-like structured membranes, there is a dense skin layer on the uppermost surface of the membrane. The bottom surface of pristine CA has smooth surface, and no pores are obvious; this led to the decrease of water flux and increase of the operation pressure [36].

As seen in Fig. 2, the presence of the extract in the membrane increases the thickness of the top dense layer while decreaseing the bottom layer. In addition, the finger-like structure becomes more elongated and larger in size. Also, enlarged cross-sectional photo displays loose sponge-like structure of the membrane. Moreover, notable pores are shown in the bottom surface of the membrane. The porosity of this modified membrane indicates that sunflower seed extract exhibited the pore-forming effect during membrane formation.

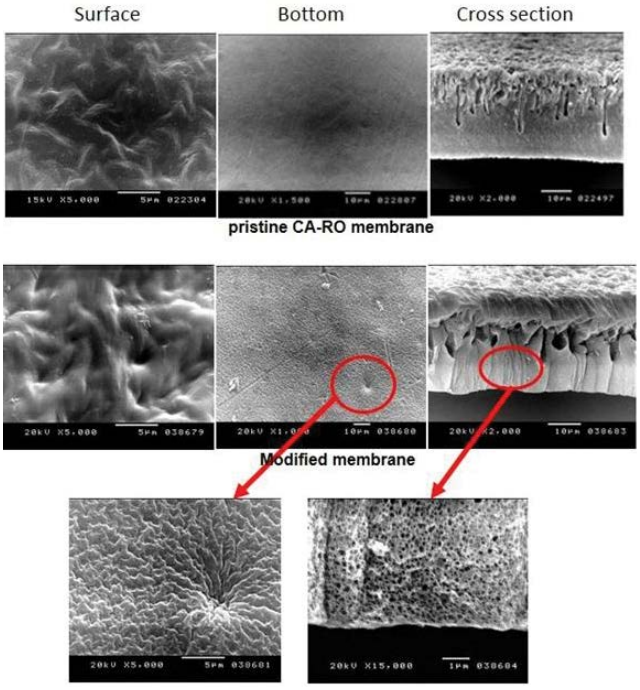

Fig. 2. SEM images of surface, bottom, and cross section of CA-RO membrane in the absence and presence of *Helianthus annuus* seed extract.

3.3. Hydrophilicity and water content

In order to evaluate the surface hydrophilicity of the pristine and modified membrane, the water contact angles of these membranes were measured and the results are presented in Table 1. It can be seen that the addition of the extract decreased the values of the contact angle. A decrease in water contact angle corresponded with the improvement in membrane hydrophilicity.

Also, the data in the table show that the water content increased by the addition of tested extract, which reveals that the addition of functionalized material enhances the hydrophilicity of the membrane and consequently improved the water content, which is well agreed with contact-angle data. This may be attributed to the existence of amine and hydroxyl hydrophilic functional groups in the membrane structures [1], which are present in functionalized extract additive. This result suggests increasing surface hydrophilicity. Moreover, the water content is also related to the porosity of the membrane, which was affected by extract addition. It is well known that the hydrophilicity of a membrane surface and the pore size and wettability of internal pore channels can greatly affect the contact angle of the membrane surface [38]. Furthermore, it is reported that lipids and proteins extracted from *Helianthus annuus* have surface active properties [35].

3.4. Salt rejection and water flux

The relation between salt rejection, water flux, and applied pressure was plotted during the desalination process (Figs. 3 and 4 and Table 2). The decrease of salt rejection with the increase of applied pressure is explained on the basis that at higher pressures, the corresponding high fluxes may lead to concentration polarization in the measuring cell, thereby causing a drop in the salt retention [39].

As seen, addition of plant extract increases the salt rejection and water flux values compared with pristine membrane up to 0.3 g of the extract. Further increase in the concentration of the extract decreases salt rejection and water flux values. The data display that 0.3 g of the extract is the optimum concentration.

The increase in salt rejection and water flux values in the presence of the additive can be explained on the basis that

Table 1
Water content and contact angle for studied membranes

Membrane	Water content, %	Contact angle
Pristine membrane	71.30	65.40
Modified membrane	75.0	53.36

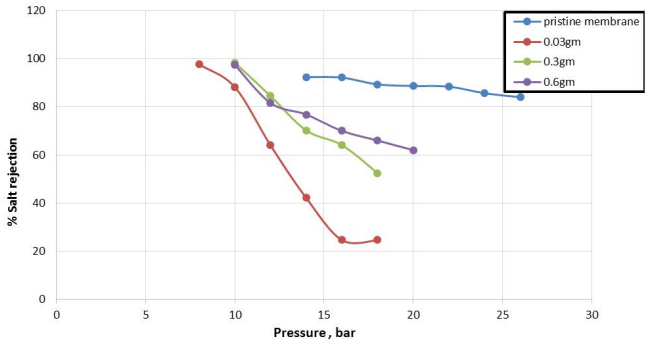

Fig. 3. The percentage of salt rejection of CA-RO membrane in the absence and presence of *Helianthus annuus* extract vs. feed pressure.

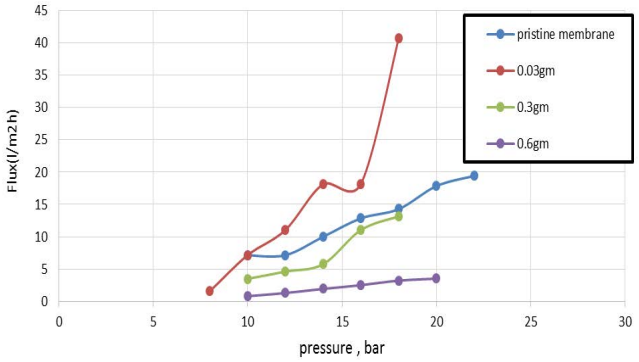

Fig. 4. Permeate water flux of CA-RO membrane in the absence and presence of *Helianthus annuus* extract versus feed pressure.

Table 2
Salt rejection percentage and water flux for studied membranes

Extract content in the membrane, g	Salt rejection, %	Water flux, L m^{-2} h^{-1}
Pristine membrane	92.3	1.14
0.03	97.7	1.56
0.3	98.4	3.48
0.6	97.5	0.78

increasing dense layer thickness with bottom layer reduction in modified membrane (as previously seen in section 3.2) improved water flux properties for these membranes. The results well agree with hydrophilicity and surface morphology data which indicated that *Helianthus annuus* seed extract increases hydrophilicity of the membrane in addition to acting as pore former. It is reported that the change in permeability of the membranes may be attributed to two possible reasons [3]. One is the increase in hydrophilicity due to the addition of the plant extract and the other is the formation of a porous membrane structure.

3.5. Antifouling properties

3.5.1. Static protein adsorption

Fig. 5 shows the protein adsorption of prepared membranes. As clearly seen, the membranes prepared with an addition of plant extract showed significantly higher resistance toward adsorptive fouling than the pristine membrane as noticed by their much lower RP%. It was found that the relative protein adsorption of pristine membrane decreased in the casting solution from 74.9 to 8.48 with the addition of the extract.

As seen, membranes containing *Helianthus annuus* seed extract have lower RP%; this result can be explained by the highest hydrophilicity of this membrane which acts as hydrophilicity modifier (section 3.3). This suggests that the hydrophilicity of membrane seemed to have an additional impact on RP%. As shown, the pristine membrane is less hydrophilic than modified membranes; therefore, the expulsion of water molecules allowed protein molecules to be adsorbed on the membrane surface [3,40]. The modified membrane surface contributed to the formation of hydration layers via ionic solvation of the charged groups and hydrogen bonds between the amide groups and the water molecules [41]. The hydration layers led to a strong repulsive force to protein at a specific distance and made the protein in contact with the membrane surface in a reverse manner [42]. Consequently, the protein adsorption is limited on modified membrane surfaces.

3.5.2. Fouling attachment on membrane

Fig. 6 illustrates SEM photograph of studied membranes after immersion in seawater for 15 d. It was found that the

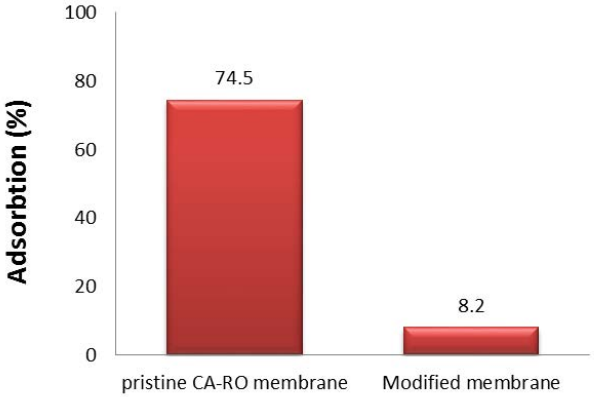

Fig. 5. Static adsorption of protein on pristine and modified cellulose acetate membrane.

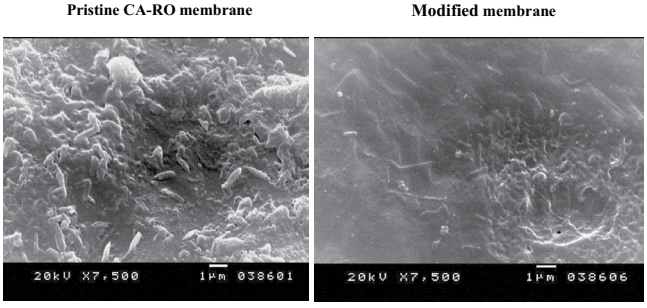

Fig. 6. SEM photograph of pristine and membrane containing *Helianthus annuus* extract after immersion in seawater for 15 d.

microbe density on modified membrane was substantially lower than that of the pristine membrane. For the membrane containing the extract, microbial attachment decreased, implying excellent anti-adhesive properties. This phenomenon may be ascribed to the presence of amine, carboxylic, or other functional groups of plant extract. Xiao-Fang et al. [43] reported that electrostatic interaction between amine, carboxylic, and other functional groups and cell membrane results in cell death [1]. According to their result, the electrostatic interaction between amine groups and carbonyl or phosphoryl groups of phospholipid components of cell membranes is the main cause of cell membrane damage in bacteria.

Also, due to the hydrophilic property of amine groups, hydrophobic materials such as proteins are expected not to be attached on them. Thus, modified membrane improved the hydrophilicity and antibacterial properties of CA membrane, which causes fewer bacteria and other hydrophobic materials to be attached on the membrane surfaces and consequently improves antifouling properties of this membrane [1]. The antimicrobial activities of *Helianthus annuus* [44] have been well documented, which may have a role in improvement of anti-biofouling properties of membranes containing this extract.

4. Conclusions

This work presents a new approach to prepare CA membranes containing aqueous extract of sunflower seeds (*Helianthus annuus*) which possess many functionalized groups in addition to its antimicrobial activity. This extract exhibited pore-forming ability during the membrane formation process and, furthermore, enhanced hydrophilicity of this membrane. Modified membrane could resist effectively protein adsorption and microbial growth. Overall, performance tests suggest that the membrane prepared with the addition of sunflower seed extract showed the best performance; therefore, they should be considered as additives in practical applications.

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