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Short communication

Synthesis, characterization and permeation performance of cellulose acetate/polyethylene glycol-600 membranes loaded with silver particles for ultra low pressure reverse osmosis

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ABSTRACT

In order to tap the seemingly boundless problem of water scarcity, reverse osmosis (RO), has been formulated as a significant solution. However, RO technology, suffers by a major challenge of biofouling, which results in reduced production capacity and increased operation costs. Thus, there is an urgent demand to fabricate an appropriate disinfection membrane surface with maximum permeation performance. In this work, cellulose acetate/poly ethylene glycol membranes impregnated with silver particles were prepared by 2-stage phase inversion protocol. The Modified membranes were characterized for their compositional analysis, surface roughness, surface morphology, permeation properties, membrane hydraulic resistance and antibacterial activity. The presence of functional group was determined by FTIR spectra. The asymmetric surface morphology of membranes was elucidated from the scanning electron microscope. The varying nature of nodules and interstices spaces was observed in the images obtained by the atomic forced microscopy. The progressive decreased was observed in the surface roughness of membranes. The modification has significantly increased the flux and salt rejection capacity of membranes. All modified membranes exhibited remarkable antibacterial properties against gram negative Escherichia Coli. The synthesis of cellulose acetate membrane, doped with polyethylene glycol and modified with silver, provides a convenient way for the development of self-sterilized membranes.

1. Introduction

Earth, often called Blue Planet, is unique among other celestial bodies because of the presence of water, which occupies two third of the earth's surface [1]. But threats of increasing water scarcity have been roused by every passing year. Currently, many of the developing countries are facing severe water scarcity, due to the rapid population growth, an advancing economy and climatic changes [2–4]. The demand for freshwater has made various areas of the world highly water-stressed, and future indicators point to escalate water scarcity throughout the world [5]. According to statistics, 1.2 billion people lack access to safe drinking water, 2.6 billion have little or no sanitation and millions of people die annually from diseases transmitted through unsafe water [6]. To increase the supply of freshwater, purification of non-conventional water sources, such as seawater or ocean water is of considerable interest due to the usual immediate proximity of such water sources for areas that are highly water-stressed [7].

Desalination of sea or saline water has been practiced regularly for over 50 years and is a well-established means of water supply in many countries [8]. Two main directions have evolved the desalination technology, namely evaporation and membrane techniques. Among various membrane processes, reverse osmosis (RO) is the most prevalent processes as it offers multiple advantages such as low energy consumption, environmental friendliness, simplicity, elevated recovery rate and high salt rejection [9,10].

The Conventional protocol used for the synthesis of membrane involves phase inversion. These processes rely on the phase separation of polymer solutions producing a range of membranes, from very porous structures to dense one [11]. The phase inversion approach involves various methods such as diffusion-induced phase separation, vapor-phase precipitation, and phase inversion by controlled evaporation and thermal-induced phase separation (TIPS).

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All the methods include complex multi-component mass transfer except thermal-induced phase separation which is primarily dependent on heat transfer [12,13].

Biopolymers have attracted much interest in the application of salt removal as they are biodegradable. Cellulose is an abundant and renewable resource found in most parts of the world, which makes it a cheap raw material for various applications [14]. It has been studied extensively in the past two decades for possible application in mopping up toxic heavy metals from the water. However, it is not effective in its natural form due to limited adsorption sites and low stability. Cellulose acetate (CA) is a well-known derivative of cellulose produced either by heterogeneous or homogeneous acetylation of cellulose [15]. CA is widely used as an RO membrane due to their good transport characteristics, low protein adsorption, appropriate mechanical strength, excellent water affinity, high hydrophilicity, excellent film-forming properties and higher desalting nature [16].

The major problem associated with cellulosic membrane is its high susceptibility to microbial attack resulting in biofouling [9]. The biofouling cannot be reduced by pretreatment alone, because deposited microbial cells can grow, multiply and relocate. Even if 99.99% of all bacteria are eliminated by pre-treatment, a few surviving cells will enter the system, adhere to surfaces, and multiply at the expense of biodegradable substances dissolved in the bulk aqueous phase [17]. Therefore, membrane biofouling has been found to occur extensively on RO membranes even after significant pretreatment of the influent stream and the addition of disinfectants such as chlorine. The sterilization with sodium hypochlorite is carried out constantly or intermittently to prevent the microbial degradation of the membrane. However, sodium hypochlorite may produce byproducts such as carcinogenic trihalomethane [18].

Incorporation of antimicrobial materials into membranes offers an innovative potential solution to biofouling control. For millennia, people have taken an advantage of the antimicrobial properties of silver (Ag). The silver ions have been studied for a wide variety of water treatment processes, including water filtration membranes [19].

In this paper, a detailed study towards the synthesis of environmentally benign membranes is reported. These membranes were prepared by using cellulose acetate, polyethylene glycol with varying amount of silver, an antimicrobial agent. A 2-stage phase-inversion protocol was devised involving thermal-induced phase inversion followed by a controlled evaporation procedure. The prepared membranes were characterized for their compositional analysis, surface morphologies, surface roughness and antimicrobial properties by using the Horizontal attenuated total reflectance-Fourier transform infrared spectroscopy (HATR-FTIR), scanning electron microscopy (SEM), atomic forced microscopy (AFM) and antibacterial assay respectively. The permeation performance, such as pure water flux, solute rejection and membrane permeability was evaluated using the reverse osmosis process.

2. Materials and methods

Cellulose acetate (CA, M_w 30,000 and acetyl content 39%), polyethylene glycol-600 (PEG), acetone and silver nitrate ($AgNO_3$) were supplied by BDH laboratories supplies Poole, England. All chemicals and solvents were used as received.

2.1. Preparation of doped solutions

8 g of CA was dissolved in 80 mL of acetone with constant stirring at 80 °C for 2 h. To this homogenous solution, 2 g of PEG was added with regular stirring at 80 °C for 6 h. The viscous and clear solution was obtained which was termed as a blended doped Solution. [20]. The casting solutions were allowed to cool down to room temperature (25 °C) and kept for 24 h in a sealed flask to remove micro bubbles formed in the solution [21].

2.2. Casting of membranes

The doped Solutions were spread slowly on a glass plate ensuring uniform thickness. The Temperature of the casted membranes was lowered to 0 °C to induce thermally induce phase separation (TIPS) which caused the formation of dense asymmetric structure. It was followed by precipitation under controlled evaporation by increasing temperature up to 60 °C [22]. The skinned membranes were obtained [23–25] which were carefully removed from the glass plates by using a sharp knife. The thickness of the resulting membranes was measured to be in the range of 0.05–0.2 mm. The membrane was named as Control.

2.3. Modification of doped solutions

Silver nitrate (0.5, 1.0, 1.5, 2.0 and 2.5%, w/v) was dissolved in ethanol (10 mL) and added to the blended dope solution with constant stirring for 2 h at 80 °C. The membranes (CPA-1–CPC-5) were casted and dried as mentioned previously (Section 2.2).

2.4. Experimental set up of reverse osmosis plant

The permeation experiments were carried out in the RO experimental rig using a plate and frame membrane module. The indigenously fabricated RO plant and its process flow diagram are shown in Fig. 1(a-b). The feed tank of 10 L capacity was filled with saline solution with conductance of 5 mS. The effective membrane area in contact with the feed was 0.018 m^2. The pressure during the process was increased from 100 to 500 kPa. Feed temperature was kept at 30 °C during the process. The permeation process was continued till it attained a steady state.

3. Characterization

The prepared membranes were subjected to various characterization techniques which are described as below.

3.1. Fourier transform infrared spectroscopy (FTIR)

FTIR spectra of membranes were recorded by using Shimadzu IR Prestige-21 equipped with Horizontal Attenuated Total Reflectance (HATR) kit and in the transmission mode at the wave number range 4000–400 cm^{-1}. The experiments were run with air as the background. For each spectrum 100 scans were accumulated with a resolution of 4 cm^{-1}.

3.2. Scanning electron microscopy (SEM)

The morphologies of the membranes were characterized by a JSM-6480, Jeol field emission scanning electron microscope. The electron beams were sputtered on sample and images of membranes on varying resolutions were observed.

3.3. Atomic forced microscopy (AFM)

AFM images were obtained by Shimadzu SPM-9500J3. Surface roughness was observed by using contact mode with oscillating tip. The scan area was chosen as 5 × 5 μm. The values of root mean square (rms) roughness were derived from AFM images, which were obtained from the average of the values measured in random areas. The membrane surface morphology can be expressed in terms of various roughness parameters, such as:

3.3.1. Mean roughness (Ra)

This parameter represents the mean value of the surface relative to the center plane, the plane for which the volumes enclosed by the