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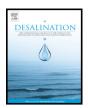
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Polyvinyl chloride and layered double hydroxide composite as a novel substrate material for the forward osmosis membrane

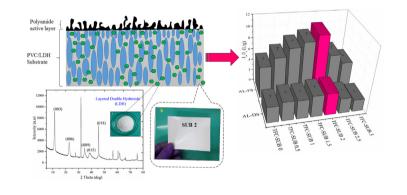
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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Novel PVC/LDH composite was used for FO substrate preparation.
- Effect of LDH on substrate morphology was studied by thermodynamic aspect.
- Effect of LDH on FO membrane performances was investigated.
- Value of *S* was able to decrease from 699 µm to 303 µm.



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ABSTRACT

In this study, a composite of the polyvinyl chloride (PVC) and layered double hydroxide (LDH) has been used as substrate material for the forward osmosis (FO) membrane preparation. Different concentrations of LDH (0, 0.5, 1, 1.5, 2, 2.5 and 3 wt%) were incorporated into PVC to fabricate FO substrates. The substrate morphology was evaluated by scanning electron microscope (SEM) and thermodynamic aspect. Furthermore, the substrate transport properties and osmotic performance of TFC-FO membranes was investigated. It was observed that the increasing concentration of LDH in PVC matrix results an increase in hydrophilicity, surface free energy, pore size and porosity of the substrates. Among all seven TFC-FO membranes, 2 wt% LDH substrate's TFC membrane (denoted as TFC-SUB 2) was unveiled a notable decrease in structural parameter (*S*). In osmotic flux evaluation, the TFC-SUB 2 membrane exhibited the high water flux of 37.46 Lm⁻² h⁻¹ (LMH) and 50.89 Lm⁻² h⁻¹ (LMH) using DI water as a feed solution and 1 M NaCl draw solution under an active layer facing feed solution (AL-FS) and an active layer facing draw solution (AL-DS) respectively. This study shows that PVC and LDH can be good candidates for the FO substrate preparation.

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1. Introduction

In recent years, numbers of large scale seawater desalination plants have been built in water stressed areas to supplement existing water

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http://dx.doi.org/10.1016/j.desal.2017.01.041 0011-9164/© 2017 Published by Elsevier B.V. resources, and it is likely to upturn in the near future. Regardless of major development in desalination technology, seawater desalination is still more energy demanding [1]. Forward osmosis (FO) is a state-of-the-art technology which uses natural osmosis to address comprehensive issues associated to water and energy [2]. In this process, osmotic pressure difference propels the water across a semi-permeable membrane from a low concentrated feed solution to a high concentrated

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draw solution. The applications of FO have been growing in many areas such as fresh water production at low energy consumption [3,4], high value solid recovery [5,6] and trace organic compounds removal [7]. Nevertheless, the concept of FO has stuck in lab scale studies due to limitations in membrane design, appropriate draw solution development and its recovery, process integration, scale up, and the feasibility of a process economy [8]. This study emphasize on membrane design, as it has been pondering a primary hindrance to FO development.

The polyamide (PA) thin film composite (TFC) membrane made-up from interfacial polymerization upon a suitable substrate is the most common commercial form of reverse osmosis (RO) and nanofiltration (NF) membrane. The main advantage of the thin film composite membrane is that interaction of both active layer and porous support layer can be individually studied to optimize overall membrane performance [9]. This concept has also been using for the FO membrane fabrication since the last two decades [9]. A TFC-FO membrane is usually composed of (i) ultra-thin polyamide active layer for high salt rejection, and (ii) highly porous support layer for water transport. A TFC membrane fabricated for FO must yield a high water flux with low salt permeability in feed and restrain the diffusion of draw solute. It must also be chemically, thermally stable and easy to manufacture at large scale. Most importantly, the FO membrane must be designed to have low structural parameter (*S*) to mitigate the osmotic inefficiency.

In the consequence of the internal concentration polarization (ICP), the importance of design of substrate in the FO membrane has conceded by many FO membrane scientists [10–12]. It does not only provide mechanical support to the polyamide active layer but also act as a suitable surface for the formation of defect free polyamide active layer [13]. To enhance the influence of the osmotic driving force more effectively, the improvement of substrate plays a crucial part in optimizing TFC-FO membrane performance.

Despite of using different polymers to tailor FO substrate, the properties like hydrophilicity, thermal, chemical, biological and mechanical stability are still big questions to achieve. The cellulose acetate, cellulose triacetate, polysulfone (PSf) and polyether sulfone (PES) are most widely used polymers for FO substrate preparation. The hydrophilic nature of cellulose acetate and cellulose triacetate can reduce ICP and increase water flux. But poor resistance to hydrolysis and biological attack makes cellulose acetate and cellulose triacetate deprived for a wide range of FO application [14]. The PSf and PES are synthetic polymers known for good chemical resistance, good mechanical properties, thermal oxidative resistance, and resistance to hydrolysis and industrial solvent. However, hydrophobic nature and a high cost of PSf and PES make them unfavorable for the FO process. In order to enhance the desirable properties of FO substrate, the incorporation of inorganic particles in polymer matrix has been attracting more attentions. It has unlocked the possibilities of making new material by combining inorganic particles into virgin polymer and also validated its performance in terms of permeability, thermal stability, selectivity, mechanical strength, and hydrophilicity [15]. Whereas, this approach has been accepted for all pressure driven membrane making process [16-18]. A varied range of polymer nanocomposites have been used to design FO porous support i.e., polysulfone (PSf)/Zeolite [19], polyether sulfone (PES)/carbon nanotube [20], polysulfone (PSf)/TiO₂ [21], polysulfone (PSf)/graphene oxide [22], polysulfone (PSf)/silica [23], polyether sulfone (PES)/reduced graphene oxide (rGO) modified graphitic carbon nitride (g- C_3N_4) [24] etc.

Regardless of increased efforts to tailor highly stable FO substrate, most of the fabricated polymer nanocomposite suffers with particle agglomeration, and non-uniform dispersion of particles. One of the possible approaches to improve the dispersion of particles in polymer matrix is applying specific functional group by surface modification of particles. The modified particles usually possess specific functional group to restrain the agglomeration in the non-polar polymer solution. Nevertheless, the complex functionalization process and deficiency of thorough understanding of the effect of functionalize particles on FO substrate limits the applications of the membrane. Hence there is need to explore a different polymer-inorganic particle composite which could improve the FO substrate quality with low cost.

Among all polymers for membrane preparation, polyvinyl chloride (PVC) has been drawing more attention as capable membrane material for its applicable mechanical, chemical resistance, film forming property, excellent thermal stability and easy to modify [25]. The PVC is very low cost material compared with other FO membrane material such as polysulfone (PSf), polyether sulfone (PES), polybenzimidazole, polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN). The PVC is easily dissolved in a many organic solvent such as *N*,*N* dimethylacetamide (DMAc), dimethylformamide (DMF), *N*-methyl-pyrrolidinone (NMP), and tetrahydrofuran (THF), which sorts it suitable for industrial membrane separation application. In recent years, numerous studies have been focused on the synthesis of ultrafiltration membrane with PVC [26,27]. However, there is need to add hydrophilic filler in to PVC matrix to increase porosity and wettability of the membrane.

Layered double hydroxides (LDHs) are highly functionalize and crystalline minerals with a positively charged brucite layer of mixed metal hydroxide [28]. LDH have many applications such as polymer additive, elimination of environmental hazardous and medical use etc. [28]. Lavered double hydroxides (LDHs) comprise of two types of metallic cations such as Mg²⁺ and Al³⁺ surrounded by a close packed configuration of OH⁻ groups in a positively charged brucite like layer, with charge compensated by anions and water located in the interlayer space [28]. This exceptional type of structure and high surface energy of LDHs provide a prodigious potential for making polymer composite material [29]. The LDH has been used as filler for a fabrication of ultrafiltration and nanofiltration composite membrane to enhanced hydrophilicity, water flux, rejection of solute and antifouling properties [30]. Furthermore, LDH can be used in a wide range of health applications and also suitable excipients in the pharmaceutical formulation [31]. Whereas, carbon base nonmaterial and graphene base nonmaterial can be a toxic for human and environment [32,33]. To the best of our knowledge, PVC/LDH composite has never been explored earlier as TFC-FO substrate material for FO.

In this study, we present a PVC/LDH composite as new material for FO substrate preparation. Our approach is to employ the properties of PVC and LDH composite for obtaining stable material to fabricate hydrophilic PVC/LDH substrate. The objectives of this study are to (i) explore the effect of LDH particles concentration in the PVC matrix on morphology by SEM and thermodynamic aspect; (ii) investigate the transport properties of the flat-sheet FO substrate; (iii) evaluate the mass transport properties and osmotic flux performance of the prepared TFC-FO membrane.

2. Materials and methods

2.1. Materials

If else specified, all chemicals and reagents were analytical grade and used without further purification. For the synthesis of LDH, magnesium chloride hexahydrate (MgCl₂·6H₂O), aluminum chloride hexahydrate (AlCl₃·6H₂O), NaOH and Na₂CO₃ were purchased from Fisher Scientific. Poly (vinyl chloride) PVC (Mw: ~48,000) (CAS No. 9002-86-2), *N*,*N* Dimethylformamide (DMF), Trimesoyl chloride (TMC), *m*-phenylenediamine (MPD), and n-hexane were purchased from Sigma-Aldrich for thin film composite membrane preparation. Polyester woven fabric sheet was supplied by Suneeta Garments (Mumbai, India). Sodium chloride (NaCl) purchased from FINAR reagent. For all experiments, deionized water (DI) (pH 5.9 \pm 0.2 and conductivity 1.0 (µS/cm)) was used (MilliQ, Millipore, India).

2.2. Preparation of LDH

The co-precipitation method was used to prepare LDH [34]. A 100 mL solution of $0.75 \text{ M MgCl}_2 \cdot 6H_2O$ and 0.25 M AlCl_3 . $6H_2O$ (molar

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