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An integrated statistic and systematic approach to study correlation of synthesis condition and desalination performance of thin film composite membranes

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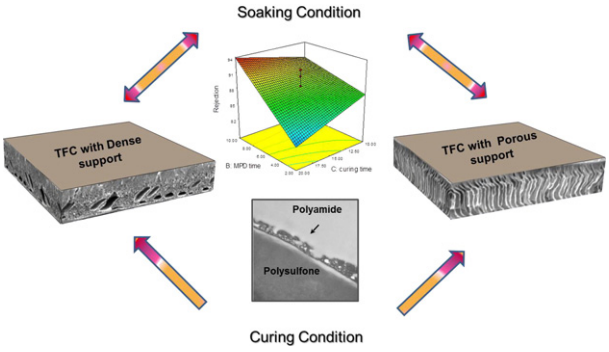
HIGHLIGHTS

- Systematic study of TFC membranes synthesis factors and their interactions.
- Statistical correlating of some synthesis variables to permeate flux and salt rejection.
- Elucidate structure-performance relationship for TFC membranes.
- PSF concentration and MPD soaking were the most significant factors on responses.
- Interaction of soaking time with curing time & PSF content on salt rejection was significant.

GRAPHICAL ABSTRACT



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ABSTRACT

Thin film composite (TFC) membranes are the most commonly used polymeric membranes in desalination process. The membrane properties and performance are dominated by many fabrication parameters, including chemistry of polymers and synthesis conditions. This study was aimed to develop an integrated statistic-systematic approach to study the correlation between fabrication condition and desalination performance of the TFC membranes. Three key fabrication factors: polysulfone concentration, aqueous phase soaking time and heat curing time were statistically analysed to determine their optimal values and interactional influence on desalination performance. The desalination performance was systematically evaluated based on permeate flux and salt rejection. It is expected that statistic results can facilitate the understanding of synthesis-performance correlations of TFC membranes. Two mathematical models for correlating fabrication factors with permeate flux and salt rejection were developed. The most influential correlations were identified as the polysulfone concentration - permeate flux and soaking time - salt rejection. Interaction of soaking time with polysulfone concentration and curing time was determined to have significant impact on the salt rejection. Our findings highlighted the importance of considering interactions between the fabrication conditions and desalination performance when devising a strategy to fabricate TFC membranes for maximising desalination performance.

1. Introduction

The development of thin film composite (TFC) membranes is the most significant achievement in the advancement of membrane

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technology for industrial applications. The TFC membranes have promising advantages in terms of desalination performance, such as high salt/organic rejection, excellent operation stability in a wide range of temperature (0–45 °C) and pH (1 − 11), and more resistance to compaction and attack by biological agents compared with conventional cellulose acetate membranes [1–3]. Therefore, the development of the TFC membranes has attracted plenty of research attention over the past decades. Typically, the TFC membranes consist of three layers: (1) a woven or nonwoven fabric layer on the bottom acting as the mechanical support to withstand high pressure operation; (2) a microporous support membrane which is directly cast on the fabric through phase inversion process; and (3) an ultrathin selective barrier layer laminated on the denser side of the microporous support by interfacial polymerization [4]. Nowadays, the most commonly used materials for the microporous support membrane and the selective barrier layer in the TFC membranes are polysulfone (PSF) and cross-linked polyamide (PA), respectively [5]. The key factors influencing the phase inversion process associated with TFC membranes' fabrication are casting solution concentration and composition, coagulation bath composition, operating conditions (temperature, humidity, etc.), and casting operations such as casting speed, film thickness and fabric pre-treatment. For the interfacial polymerization of selective barrier layer, the most important parameters include the type and concentration of monomers, additives, organic solvents, soaking and reaction times, curing time and temperature and washing strategies [1,2,6]. The overall desalination performance of the TFC membranes could be improved progressively through manipulation of these fabrication factors and optimization of physiochemical characteristics of individual layers in terms of their resistance, efficiency and formation [3,7].

To date, there are many studies on identifying key fabrication parameters and their impact on desalination performance of the TFC membranes [8–21]. Even though a number of studies have been conducted to assess the influence of the fabrication parameters, interaction of the selected factors appears to be poorly investigated systematically. For instance, some studies were conducted to investigate the effect of microporous support structure and chemistry of the TFC membranes on their performance [5,20,22]. They found that the support layer characteristics have profound influence on the separation performance. However, the fabrication parameters of PA selective layer made with different support structures were fixed in these works. Other reports documented the correlation between the filtration performances and curing time associated with interfacial polymerization, suggesting that curing time has significant influence on TFC membranes' performance [6,9,10]. Another factor which is believed to be important for the performance of TFC membranes is the aqueous phase soaking time. However, correlation between the soaking time and desalination was varied in the reported studies [17,18,20]. Most of previous studies only focused on one-on-one correlation between fabrication factors and filtration performance of the TFC membranes. It is believed that these factors, aqueous phase soaking and curing time and PSF structure, should have some interactions, which can also affect the filtration performance. A better understanding of possible interactions between fabrication factors and desalination performance of the TFC membranes could be beneficial for the optimization of the fabrication processes. Furthermore, statistical methods can be a useful tool for studying the interactions and identifying the optimum separation performance [7,17].

Herein, this study was aimed to develop integrated statistic-systematic analysis approach together with Design Expert software to identify interactions of fabrication variables and their correlations with the desalination performance of TFC membranes. Polysulfone concentration, aqueous phase soaking time and heat curing time were selected as key fabrication factors, as which are believed to have predominant influences on mechanical property and desalination performance of the TFC membranes [1,2,6]. It could be hypothesised that these factors could have interactions within the fabrication process and with the filtration performance, which are worth studying. To our knowledge, no reports were found in the literature studying how the aqueous phase soaking time influences on the resulting TFC membranes' performance considering the variations in curing time and support layer porosity. TFC membranes were fabricated following a two-step casting method by fixing fabrication parameters other than the selected independent factors at the most common or optimum reported values (referred in Section 3.1). The obtained TFC membranes will be evaluated in terms of permeate flux and salt rejection using a cross-flow filtration unit. We will use full factorial design to design the experiments and statistical methods to establish mathematical models expressing the fabrication factor-desalination performance correlations. Desirability function approach (DFA) is finally used for multiple response optimization [23]. The research outcomes will provide better insights into the interrelationship among these variables, which can be helpful for simultaneously optimizing the synthesis stages of TFC membranes and their desalination performance.

2. Materials and methods

2.1. Materials

Commercial polyester non-woven fabric (100 μm) was supplied by PURUIXIN-TOP-SCIENCE and used as the backing layer for PSF interlayer. PSF (Udel P-1700 CL2611 CMP, Mw ≅ 45,000) was provided by Solvay Speciality Polymers and used to fabricate PSF support interlayer. *N*,*N*-dimethylformamide (DMF, anhydrous, 99.8%, Merck) was used as the solvent to dissolve PSF. An adjustable casting knife (Elcometer 3530/2, Elcometer, UK) was used for casting PSF support layer on glass surface. For the establishment of PA selective layer on the PSF support. 1,3-Phenylenediamine (MPD, >99%), 1,3,5-benzenetricarbonyl trichloride (TMC, 98%), and *n*-hexane were purchased from Sigma Aldrich. Sodium chloride (NaCl) was purchased from VWR International Company.

2.2. TFC membrane fabrication

The TFC membranes were synthesized through a two-stage casting method following a full-factorial design. For the support layer, PSF membrane was made through phase inversion process at different PSF concentrations (9–15% (w/w)). Typically, PSF beads were dried at 60 °C overnight and added to DMF. The mixture was stirred at room temperature to form a clear homogenous solution and then degassed in an ultrasonication bath equipped with degassing function for 3 h. Subsequently, the non-woven fabric was placed on a clean glass plate and wetted with degassed DMF. Excessive solvent was removed by a soft rubber roller on the surface of fabric followed by tapping fabric to glass plate. After that, the above-prepared PSF solution was casted on the polyester fabric using an adjustable casting knife with a fixed height at 200 μm. The glass plate was then immediately immersed in a precipitation bath containing distilled water and 2% DMF at 22 °C to initiate phase inversion process. The precipitated PSF supports were allowed to stay in the bath for at least 10 min to ensure complete phase separation and then washed thoroughly with distilled water and stored in distilled water at 5 °C for further experiments. In order to fabricate the PA selective barrier layer of TFC membranes, the synthesized PSF support was clamped between an acrylic plastic plate, a rubber gasket and an acrylic plastic frame on the top of that. Then, 2% (w/v) MPD aqueous solution was poured inside the frame and allowed to wet the PSF support for 2–10 min based on the designed experiments. After lapsing the designated MPD soaking time, excess MPD solution was drained. Then the frame was disassembled and extra MPD was removed using a soft rubber roller pressed on PSF until no visible droplets were remained on the surface of the PSF support. The plate-gasket-frame disassembly/rolling/reassembly period was fixed at 2.5 min in all experiments. Following to that, 0.1% (w/v) TMC in n-hexane solution was poured into the frame and allowed to react with the adsorbed MPD for 1 min resulting in the