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Design and fabrication of inner-selective thin-film composite (TFC) hollow fiber modules for pressure retarded osmosis (PRO)

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ABSTRACT

Pressure retarded osmosis (PRO) is a promising technology to harvest the renewable osmotic energy from salinity gradients. There are great progresses in the fabrication of PRO membranes in the last decade. Thin-film composite (TFC) hollow fibers have been widely studied and demonstrated superior performance. However, the lack of effective TFC hollow fiber modules hinders the commercialization of the PRO technology. Knowledge and experiences to fabricate TFC hollow fiber modules remain limited in the open literature. In this study, we aim to reveal the engineering and science on how to fabricate TFC hollow fiber modules including the formation of inner-selective polyamide layers and the repair of leakages. TFC-PES hollow fiber modules with 30% and 50% packing densities have been successfully fabricated, showing peak power densities of 20.0 W/m^2 and 19.4 W/m^2, respectively, at 20 bar using 1 M NaCl solution and DI water as feeds. The modules may be damaged during handling and high pressure testing. The repaired modules have a power density of 18.2 W/m^2, 91% of the power densities of the undamaged ones. This study would make up the gap between TFC membrane fabrication and TFC membrane module fabrication in the membrane industry.

1. Introduction

Exploits of renewable energies are essential to meet the escalating energy demands. Osmotic energy from salinity gradient, with a projected magnitude of 2.6 TW, is one of the promising sources of sustainable energies [1–6]. Several processes, such as pressure retarded osmosis (PRO) [6], reverse electro-dialysis (RED) [7], capacitive mixing [8] and hydrogel swelling [9], are proposed to harvest the osmotic energy. PRO is the most widely investigated process because of its greater efficiency, higher power density and potential integration with reverse osmosis (RO) plants to lower the energy consumption for seawater desalination [1–4,6,10]. In PRO, water spontaneously permeates from a low-salinity feed solution through a semi-permeable membrane to a high-salinity draw solution against an applied hydraulic pressure difference [10–12]. The resulting pressurized diluted salty water with an expanded volume will go through a hydro-turbine [11–15] or a pressure exchange [16–18] to release its osmotic energy in useful forms.

As the heart of PRO processes, PRO membranes have received great attentions and significant progresses have been made since Loeb and Mehta tested commercial RO membranes for PRO applications in the 70 s [11,12,19–21]. In the last decades, thin-film composite (TFC) membranes gained increasing considerations due to their higher water fluxes and higher power densities [5,22–25]. Most TFC membranes are formed in-situ onto the surface of microporous substrates via interfacial polymerization of aromatic diamine such as piperazine (PIP) [26] and m-phenylenediamine (MPD) [27] with acid chloride monomers such as trimesoyl chloride (TMC) [28] and isophthaloyl chloride (IPC) [29]. Hydration Technology Innovations (HTI) has developed a commercial TFC flat-sheet membrane, which was widely tested in the literature [30–32]. In comparison to TFC flat-sheet membranes, TFC hollow fiber membranes have a higher surface area per volume, a self-mechanical support and no shadow effects [6,33]. In spite of their advantages, there are no commercially available TFC hollow fiber membranes or modules due to its complicated fabrication process. Up to date, the only commercially available PRO hollow fiber membrane is a cellulose triacetate (CTA) membrane developed by Toyobo. It has a reported power density of 10.1–13.5 W/m^2 using 1.2 M seawater reverse osmosis brine and freshwater as the feed pair [34–36].

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In our previous studies, we developed a robust inner-selective TFC polyethersulfone (PES) hollow fiber membrane that exhibited a power density above 25 W/m^2 using 1 M NaCl solution and DI water as the feed pair at 20 bar [37,38]. This membrane must be assembled into useful forms, i.e., modules, in order to modularize installation and operation and fully access the feasibility of PRO [39–41]. Therefore, module engineering plays an important role to optimize membrane performance for PRO in terms of increasing the membrane area per module, optimizing the flow pattern and enhancing the mechanical properties of the modules [39,42].

However, techniques to fabricate hollow fiber modules have always been kept as trade secrets. Up to date, there are limited literatures on fabrication of scalable hollow fiber modules [43–46]. In most studies, interfacial polymerization was conducted on mini-modules comprising 3–10 pieces of hollow fiber membranes instead of scalable modules [40,41].

In this work, we investigated the science and engineering to fabricate TFC hollow fiber module by assembling hollow fibers into a semi-pilot-scale module and subsequently conducting interfacial polymerization to form thin-film layers on the inner surfaces of the hollow fibers. These knowledge and skills are important to fabricate TFC hollow fiber modules for PRO, reverse osmosis, nanofiltration and other applications, bridging the gaps between membrane fabrication and module fabrication.

2. Experimental

2.1. Materials

Radel® A polyethersulfone (PES, Solvay Advanced Polymer), N-methyl-2-pyrrolidone (NMP, >99.5%, Merck), polyethylene glycol 400 (PEG, Mw = 400 g/mol, Sigma-Aldrich) and deionized (DI) water were used as the polymer, solvent and non-solvent additives, respectively, to prepare the polymer dope of the hollow fiber substrates. A 50/50 wt% mixture of glycerol (Industrial grade, Aik Moh Pains & Chemicals, Singapore) and DI water was employed for the post-treatment of as-spun hollow fiber substrates. *m*-phenylenediamine (MPD, >99%, Sigma-Aldrich), sodium dodecyl sulphate (SDS, >97%, Fluka) and DI water were acquired to prepare the MPD solution; Trimesoyl chloride (TMC, >98%, Tokyo Chemical Industry, Japan) and hexane (>99.9%, Fisher Chemicals) were utilized to prepare the TMC solution for the interfacial polymerization. Epoxy (KSbond, Kuo Seng Enterprise, Taiwan) was purchased to cast the tubesheets of the hollow fiber modules.

2.2. Fabrication of the TFC-PES hollow fiber membranes

The PES hollow fiber substrates were prepared by a dry jet wet spinning process [47]. The polymer dope composition and spinning conditions have been reported previously [37,48]. Minor modifications were made to further enhance the mechanical properties and PRO performance. The modified spinning parameters are presented in Table 1. The outer and inner diameters of the hollow fiber membrane are 575 μm and 1023 μm, respectively. The as-spun hollow fiber substrates were collected from the spinning line and soaked in water for 2 days to remove the residual solvent and non-solvent additives. The PES hollow fiber substrates were then posted in a 50/50 wt% glycerol/water solution for another 2 days. After drying, three pieces of the PES substrates were assembled into a mini-module. The TFC-PES hollow fiber membranes were then formed via interfacial polymerization of MPD and TMC on the inner surfaces of the hollow fiber membranes [37,47].

Table 1
Dope composition and spinning conditions of the PES hollow fiber supports.

Spinning parameters	PES membrane substrate
Bore fluid	Deionized water
Dope composition (wt%)	20–22/36–39/36–39/1–4 (PES/PEG/NMP/Water)
Outer channel	NMP
External coagulant	Water
Outer flow rate (ml/min)	0.03–0.1
Inner dope flow rate (ml/min)	2.0–3.0
Bore fluid flow rate (ml/min)	0.8–1.2
Air gap length (cm)	1–3
Take up speed (m/min)	1–5
Dual layer spinneret dimension (mm)	Outer OD = 1.6 mm, inner OD = 1.3 mm, ID = 1.14 mm

2.3. Fabrication of TFC-PES hollow fiber modules

Fig. 1 shows a general configuration of the inner-selective PRO hollow fiber module with the draw solution flowing along the lumen side and the feed solution flowing along the shell side. The housing has two ports for the introduction and exit of the feed solution, respectively. The draw solution enters the port from one end cap, flows though the fiber lumens and exits from the other end cap. Tubesheets are formed at the two ends of the housing with openings to the lumens of the hollow fiber membranes. The tubesheets work as physical barriers to separate the feed solution from the draw solution and prevent mixing. Therefore, the only path for the feed solution to meet the draw solution is by passing through the walls of the hollow fiber membranes.

2.3.1. Fabrication of PES hollow fiber modules

The step-by-step fabrication of a hollow fiber module is presented in Fig. 2. The PES hollow fiber substrates after the glycerol post-treatment were collected into a bundle. The amount of hollow fibers in each bundle was calculated to meet the target packing density, which was defined as the volumetric fraction of the modules occupied by the hollow fibers.

$$n = \frac{\phi D^2}{d^2} \quad (1)$$

where n is the total number of hollow fibers in the module, ϕ is the packing density, D is the inner diameter of the module housing and d is the outer diameter of the hollow fiber.

Hollow fiber modules with 30% and 50% packing densities were prepared. The simplest way to prepare the hollow fiber bundle is to collect the hollow fibers and arrange them in parallel randomly to form a bundle. However, when the packing density is low, the hollow fiber may not distribute uniformly and fill up the housing [49]. To overcome these shortcomings, structured hollow fiber bundles were prepared by laying down the hollow fibers in parallel at a uniform spacing to form a hollow fiber mat and then rolling up the mat to form a structured bundle [50,51]. This method is relatively complicated but yields a more uniformly packed bundle

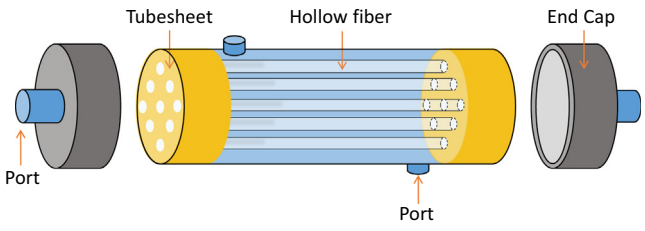

Fig. 1. Schematic drawing of a PRO hollow fiber module.