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Helical hollow fibers via rope coiling: Effect of spinning conditions on geometry and membrane morphology

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ABSTRACT

Helical hollow fiber membranes from poly(ether sulfone) were spun via dry-wet spinning, making use of the liquid rope coiling phenomenon. The polymer solution composition was changed by varying the coagulation value and adding PEG400 as pore former. The bore liquid composition, outer coagulation bath temperature, air gap, polymer dope and bore liquid flowrates were varied to map the conditions where helical fibers form. It was observed that increasing air gap changed fiber geometry from straight to helical. Increasing the outer coagulation bath temperature caused helical geometries to become irregular or straight under identical spinning conditions, possibly due to the higher water vapour absorption in the air gap. Regularly helical fibers could, however, be spun with a coagulation bath at 50 °C under a variety of conditions. Comparing fibers spun under the same conditions with different solutions, it was observed that as the solution's viscosity increased, the geometry shifted towards helical. Increasing the bore liquid solvent strength so as to form the skin on the bore side yielded straight fibers due to the fast solidification of the fibers before curling and all helical fibers fabricated had a denser skin on the shell side. During filtrations of yeast suspensions, the helical fibers experienced less fouling with the feed on either the bore or the shell side of the membrane, which shows that using liquid rope coiling to spin helical fibers is a promising, practical method of alleviating fouling in membrane filtrations.

1. Introduction

Concentration polarization and membrane fouling are among the most significant bottlenecks in pressure-driven membrane processes, as they decrease membrane productivity and lifetime. There are numerous studies and many different methods of attack to alleviate the effects of fouling. Among these methods, one that has been attracting increasing interest lately, due to the advances in micro- and nano-fabrication techniques, is the introduction of patterns on a membrane surface so as to produce vortices that enhance transport of rejected components away from the membrane surface. Producing vortices at low Reynolds numbers this way has the advantage of lower energy consumption and avoiding high shear rates, which may be undesired for shear sensitive materials such as proteins. Micropatterns on flat [1–10] and hollow fiber [11,12] membrane surfaces have been reported to decrease deposition on the membrane surface and improve membrane performance in microfiltration [2–4,1], ultrafiltration [5–7,11,12], nanofiltration [8,9] and membrane distillation [10] processes. In a recent study, Luelf et al. fabricated fibers with sinusoidally narrowing and widening bore channels by pulsed flow of the bore liquid and have shown that such a pattern also increases mass transfer rate in gas/liquid contacting [13].

Dean vortices are secondary flows which develop due to the imbalance between viscous and centrifugal forces when a fluid is moving in a curved duct. Dean number, De, which represents the ratio of centrifugal to viscous forces is defined as follows for flow inside a tube or hollow fiber:

$$De = Re\sqrt{\frac{D_{in}}{D_{coil}}} \tag{1}$$

Here Re is the Reynolds number, D_{in} is the inside diameter of the fibers and D_{coil} is the effective coil diameter defined as:

$$D_{coil} = D_{helix}\left[1 + \left(\frac{P}{\pi D_{helix}}\right)^2\right] \tag{2}$$

Here, D_{helix} is the diameter of the helix and P is the pitch as shown in Fig. 1.

The application of flow instabilities including Dean vortices in membrane processes for improving mass transfer dates back to the several decades ago. By winding hollow fiber membranes around a perforated tube, coiled channels were obtained and their performance was compared to straight tubular membranes in filtrations of a variety of feeds such as bentonite [14–17], yeast [18,15,19,20], activated

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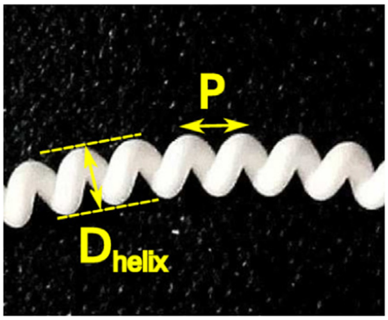

Fig. 1. Dimensions, D_{helix} and P, of helical fibers.

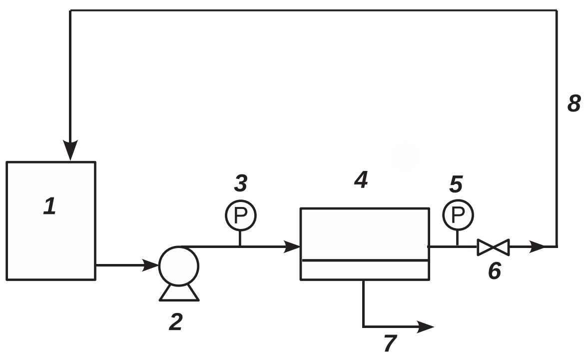

Fig. 2. Schematic of the crossflow filtration setup. 1) Feed tank, 2) peristaltic pump, 3) feed pressure gauge, 4) membrane module, 5) retentate pressure gauge, 6) back-pressure valve, 7) permeate line, 8) retentate line.

Table 1
The polymer solutions used in membrane fabrication and their viscosities (at 1 s^{-1} shear rate).

Solution	PES (wt%)	Triton X-100 (wt%)	NMP (wt%)	PEG 400 (wt%)	Water (wt%)	Coagulation value	Viscosity (Pa.s)
M	15.3	5.1	72.3	–	7.3	75%	1.79
N	15	5	70.7	–	9.3	95%	2.48
O	15	5	52.4	20	7.6	95%	5.49

sludge [15], bovine serum albumin [19], dextran [14,19], natural waters [17], salts [21,22] and amino acids [21]. A number of studies [23,24,16] have compared coiled, woven and meandering geometries, which all generate Dean vortices, during membrane filtrations. Ghogomu et al. have reported that all curved geometries gave the same limiting flux during bentonite filtration [16], whereas Kuakavi et al. have concluded that woven fibers consumed less energy compared to helical ones in the filtration of dextran solutions [23]. Luque et al. have shown that coiled modules offer flux and capacity improvement in both constant pressure and constant flux filtrations of yeast suspensions at different concentrations [20]. Kaufhold et al. have stated that mass transfer rates can only be improved at small curvature diameters less than 4 mm and that helical and twisted fiber geometries would only be technically feasible with respect to module costs if these are produced directly during the spinning process [24]. In all these studies, it was shown that geometries forming Dean vortices resulted in improved performance with less foulant deposition and higher permeate fluxes, which eventually required less energy consumption for the same filtration.

While it is possible and quite effective, as shown by the mentioned studies, to construct coiled hollow fiber membranes from originally straight ones, it is more practical and less costly to produce coiled hollow fibers during the fiber spinning process. The process of forming helical fibers during spinning relies on the natural phenomenon called

Table 2
Spinning conditions of the hollow fiber membranes.

Membrane	PDFR (mL/min)	BLFR (mL/min)	PDFR/BLFR	Bore liquid	Air gap (cm)	Coagulation bath temperature
M1	8.61	2.59	3.3	80:20	4	20 °C
M2	8.61	2.59	3.3	80:20	6	20 °C
M3	8.61	2.59	3.3	80:20	8	20 °C
M4	11.48	3.45	3.3	80:20	4	20 °C
M5	11.48	3.45	3.3	80:20	6	20 °C
M6	11.48	3.45	3.3	80:20	8	20 °C
M7	17.23	5.18	3.3	80:20	8	20 °C
M8	8.61	2.59	3.3	80:20	8	50 °C
M9	11.48	3.45	3.3	80:20	8	50 °C
M10	18.18	5.18	3.5	80:20	10	50 °C
M11	18.18	5.18	3.5	70:30	10	50 °C
M12	18.18	3.45	5.2	70:30	10	50 °C
M13	18.18	1.73	10.5	70:30	10	50 °C
M14	18.18	0.85	21.4	70:30	10	50 °C
M15	18.18	5.18	3.5	70:30	8	50 °C
M16	18.18	3.45	5.2	70:30	8	50 °C
M17	18.18	1.73	10.5	70:30	8	50 °C
M18	18.18	0.85	21.4	70:30	8	50 °C
N1	18.18	5.18	3.5	80:20	8	50 °C
N2	18.18	5.18	3.5	80:20	10	50 °C
N3	18.18	3.45	5.2	80:20	10	50 °C
O1	11.48	3.45	3.3	80:20	10	50 °C
O2	17.23	5.18	3.3	80:20	10	50 °C
O3	17.23	5.18	3.3	80:20	8	50 °C

"liquid rope coiling", a term first defined by Barnes and Woodcock in 1958 [25]. The periodic buckling, which forms the coiling fibers is observed when a viscous fluid is poured onto a solid or liquid surface from a certain height. Coiling occurs as a result of the competition between axial compression and bending of the fluid "rope", and is influenced by the fluid's viscosity, density, flow rate and radius, as well as the height and gravity. Three distinct coiling regimes have been defined by Ribe [26] depending on the relative magnitudes of viscous, gravitational and inertial forces, and each of these regimes are shown to be governed by different relationships between the coiling frequency and the mentioned parameters.

Spinning polymeric fibers via liquid rope coiling have been shown for full cellulose fibers [27] and recently for poly(ether sulfone) (PES) hollow fiber membranes [28]. Luelf et al. have fabricated helical hollow fibers and shown that increasing polymer dope and bore liquid density, increasing extrusion speed and falling height (air gap), and decreasing pulling speed favor the rope coiling effect [28].

In the current study, we thoroughly investigate the effects of fabrication parameters on the geometry and morphology of PES hollow fiber membranes. Polymer dope composition is varied to observe the effects of viscosity and coagulation value of the solution, while bore liquid composition is varied to affect the nonsolvent strength. Flowrates of polymer dope and bore liquid are varied to see the effects of acceleration of the extruded jet on coiling as well as the overall dimensions. Finally the coagulation bath temperature was changed from room temperature to 50 °C to produce vapour in the air gap in order to start vapour-induced phase separation prior to solidification in the coagulation bath. At the end, we illustrate the decreased fouling in helical fibers during filtration of yeast suspensions both in inside-out and outside-in configurations.

2. Experimental

2.1. Materials

Poly(ether sulfone) (PES, Ultrason E6020P) was supplied by BASF. N-methyl pyrrolidone (NMP, 99%), poly(ethylene glycol) (PEG 400), Triton® X-100, Bovine Serum Albumin (BSA) and phosphate-buffered