



Design and modeling of novel low-pressure nanofiltration hollow fiber modules for water softening and desalination pretreatment



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ABSTRACT

Given its high surface area to volume ratio and desirable mass transfer characteristics, the hollow fiber module configuration has been central to the development of RO and UF technologies over the past five decades. Recent studies have demonstrated the development of a novel class of low-pressure nanofiltration (NF) hollow fiber membranes with great promise for scale-up implementation. Further progress on large-scale deployment, however, has been restrained by the lack of an accurate predictive model, to guide module design and operation. Earlier models targeting hollow fiber modules are only suitable for RO or UF. In this work, we propose a new modeling approach suitable for NF based on the implementation of mass and momentum balances, coupled with a validated membrane transport model based on the extended Nernst-Planck equation to predict module performance at the system-level. Modeling results are validated with respect to synthetic seawater experiments reported in an earlier work. A preliminary module design is proposed, and parametric studies are employed to investigate the effect of varying key system parameters and elucidate the tradeoffs available during design. The model has significant implications for low-pressure nanofiltration, as well as hollow fiber NF module design and operation.

1. Introduction

Nanofiltration (NF), introduced in late 80s, is a membrane process whose performance falls between ultrafiltration (UF) and reverse osmosis (RO); and as its name implies, has pore sizes on the order of 1 nm (corresponding to a molecular weight cut-off, or MWCO, of 300–500 Da) [1]. With their unique selectivities and high permeabilities relative to RO, NF membranes presented a major milestone in membrane technology, have attracted considerable attention since their introduction, and have found numerous applications, spanning numerous fields from water and waste water treatment to biotechnological, pharmaceutical, and food industry applications [2]. A recent review by Mohammad et al. [2] identifies NF as a research domain of surging interest, predicts NF will continue to play a prominent role in membrane technology, and reveals the future prospects and areas of potential growth NF is likely to experience in the long term. Recent studies have demonstrated the development of a novel class of low-pressure NF hollow fiber membranes with great promise for scale-up implementation [3, 4, 5]. The design of hollow fiber membrane modules based on those membranes is the focus of the current work.

Given its high selectivity for monovalent ions, areas in which NF is projected to grow include hard water softening, and more recently, desalination pretreatment. While traditional softening technologies, such as lime softening and pellet softening, have been challenged by their large sludge production and chemical consumption, NF has emerged as a viable alternative [6, 7]. Apart from traditional softening, NF desalination pretreatment has lately been gaining attention as evident from a recent review on integrated/hybrid membrane processes in desalination and water treatment by Ang et al. [8]. The review cites evidence that NF desalination pretreatment not only improves desalination feeds by reducing scaling, but also allows seawater reverse osmosis (SWRO) to run at lower pressures and potentially achieve higher recoveries [8]. In search of the optimal conditions for NF/SWRO desalination, Park et al. followed with their study on NF pretreatment and its effect on SWRO recovery under different conditions, and concluded that the NF/SWRO configuration featured an improvement in the quality of recovered water, especially at high recoveries [9]. More recent works from Roy et al. [10, 11] explore the effect of temperature on NF pretreatment and scale prevention in thermal desalination.

As becomes apparent from Bergman's cost analysis of softening

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Nomenclature*Roman symbols*

| | |
|----------------|--|
| A | Debye–Hückel constant, $\text{m}^{3/2}/\text{mol}^{1/2}$ |
| A_c | flow cross-sectional area, m^2 |
| A_k | membrane porosity |
| c_i | solute concentration, mol/m^3 |
| d_i | fiber inner diameter, m |
| d_o | fiber outer diameter, m |
| CP_i | concentration polarization factor |
| $D_{i,p}$ | diffusion coefficient in the pore, m^2/s |
| $D_{i,\infty}$ | diffusion coefficient in the bulk, m^2/s |
| D_f | fractal dimension |
| D_h | hydraulic diameter, m |
| $D_{i,module}$ | module inner diameter, m |
| e | specific energy consumption, kWh/m^3 |
| e_0 | elementary charge, $1.60218 \times 10^{-19} \text{ C}$ |
| f_D | Darcy friction factor |
| F | Faraday constant, $96,485.3 \text{ C}/\text{mol}$ |
| I | ionic strength, mol/m^3 |
| J_v | permeate flux, $\text{m}^3/\text{m}^2 \cdot \text{s}$ |
| k | Boltzmann constant, $1.38065 \times 10^{-23} \text{ J}/\text{K}$ |
| $k_{c,i}$ | solute mass transfer coefficient, m/s |
| $K_{i,c}$ | convection hindrance factor |
| $K_{i,d}$ | diffusion hindrance factor |
| L | module length, m |
| N_A | Avogadro's number, $6.02214 \times 10^{23} \text{ mol}^{-1}$ |
| N_f | number of fibers in the module |
| \dot{N}_i | molar flow rate of species i , mol/s |
| P_w | wetted perimeter, m |
| Q | volumetric flow rate, m^3/s |
| r_p | effective pore radius, m |
| r_i | solute Stokes radius, m |
| R | universal gas constant, $8.31446 \text{ J}/\text{mol K}$ |
| R_{ex} | experimental solute rejection |
| R_i | modelled solute rejection |
| RR | recovery ratio |
| Re | Reynolds number |
| Sc | Schmidt number |
| Sh | Sherwood number |
| T | temperature, K |
| V | channel bulk velocity, m/s |
| X_d | membrane charge density, mol/m^3 |
| z_i | ion valency |

Greek symbols

| | |
|--------------|---|
| Δz | cell thickness, m |
| ΔP | applied pressure difference across the membrane, Pa |
| $\Delta \Pi$ | osmotic pressure difference across the membrane, Pa |
| ΔW_i | born solvation energy barrier, J |
| Δx | thickness of membrane active layer, m |
| γ_i | activity coefficient |
| λ_i | ratio of solute Stokes radius to effective pore radius |
| μ | solvent viscosity, Pa·s |
| ϕ | packing density |
| ϕ_i | ratio of permeate flux to the uncorrected mass transfer coefficient |
| Φ_i | steric partitioning coefficient |
| Φ_B | born solvation coefficient |
| ψ | electric potential, V |
| Ψ_f | correction factor for fractal dimension |
| ρ | solution density, kg/m^3 |
| ϵ_0 | permittivity of vacuum, $8.85419 \times 10^{-12} \text{ F}/\text{m}$ |
| ϵ_b | relative permittivity/dielectric constant of the bulk |
| ϵ_p | relative permittivity/dielectric constant of the pore |
| ϵ_r | relative permittivity/dielectric constant |
| ξ | electric potential gradient at the feed/membrane interface, V/m |
| Ξ_i | mass transfer coefficient correction factor |

Subscripts

| | |
|-------|-----------------------|
| b | bulk feed solution |
| D | Donnan |
| D_h | hydraulic diameter |
| f | feed |
| in | inlet |
| L | hydraulic loss |
| p | permeate |
| w | condition at the wall |

Superscripts

| | |
|-----------|---|
| \bullet | mass transfer correction for the suction effect |
| $*$ | optimal feed flow rate for a given pressure |

technologies, however, one primary limitation on the economic viability of the NF pretreatment system has been the additional energy penalty incurred by the process [7]. Consequently, the requirement of lower energy consumption has occupied membrane researchers and process designers alike in search of better separation efficacy. While most NF membranes have been thin film composite (TFC) flat sheet membranes, a novel class of low-pressure NF hollow fiber membranes developed using layer-by-layer (LbL) polyelectrolyte deposition with chemical cross-linking appears to provide a solution [3, 4, 5]. According to performance tests reported in earlier work [4, 5], the newly developed membranes fared well when compared to state-of-the-art in softening and possessed superior softening performance compared to commercial NF membranes while operating at pressures that did not exceed 5 bar. These results prove the membrane's potential, raising research interest for potential large-scale implementation.

Despite their great potential, further progress on scale-up implementation of these or other NF hollow fiber membranes is restrained by the lack of information on large-scale or commercial NF hollow fiber

modules, which is necessary to evaluate a membrane's potential for scaling-up. In this work, we develop a mathematical model to our knowledge the first to predict the performance of NF hollow fiber modules on the system-level, building from experiments run on a bench-scale setup. In an earlier study, we demonstrated the successful implementation of a membrane transport model, introduced by Geraldes and Alves [12] based on the extended Nernst-Planck equation, to the newly developed LbL hollow fiber membranes [13]. However, that model focused on coupon-sized systems, and did not take into consideration the streamwise variations that will be inherent in large-scale applications. To address this need, a new model, taking a deeper look into the fundamentals governing fluid flow and mass transfer in hollow fiber modules, becomes necessary.

Since they were first patented in 1966 by DOW Chemical Company [14] followed by DuPont [15], research and development on hollow fiber membrane modules has made significant progress. These modules became attractive from an application standpoint given their desirable mass transfer characteristics and high surface area to volume

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