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A critical review of transport through osmotic membranes

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ABSTRACT

Herein, we review mechanisms and models of solute transport relevant to nanofiltration (NF), reverse osmosis (RO), and forward osmosis (FO) membrane separation processes. We first consider state of the art polymeric NF, RO, and FO membrane properties and how these properties influence water and solute transport. Next, we critically review classical models (*i.e.*, pore flow, solution-diffusion, and Nernst–Planck) and discuss the shortcomings of each, particularly focusing on their inability to explain fundamental relationships between water/solute transport and membrane properties, such as free volume, crystallinity, extent of swelling, and the presence of filler materials in mixed matrix/nanocomposite membranes. We then review modern structure–performance models that attempt to relate water/solute transport to quantifiable membrane structure and solute/water/polymer interactions. Finally, we consider both classical and modern mass transfer models that describe external, internal, and enhanced concentration polarization – all of which strongly influence water/solute transport through NF, RO, and FO membranes.

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1. Introduction

Clean water supplies are dwindling in the modern world. While the quantity of fresh water remains relatively constant, the demand of our rapidly increasing population has exploded in the last century [1]. Inefficient use of water, climate change driven water scarcity, and industrial pollution have led to the current state in which nearly 1.8 billion people live in water scarce regions and over 780 million people lack access to improved drinking water [2]. Thanks to global efforts of improved sanitation and infrastructure access to improved drinking water has been expanded significantly in the last 20 years [2]. To ensure further improvements and maintain water security into the future new freshwater sources will need to be explored further, such as reclaimed wastewater and desalinated seawater.

Membrane processes are now the primary separation technology used in wastewater reclamation and desalination. These technologies are efficient and effective, but there is a continued need to expand and improve their capabilities [3–7]. Improved NF membranes now enable highly energy-efficient removal of color-causing organic matter, hardness minerals, toxic metals, and chemicals of concern, from both natural and industrial waters [8]. Secondary municipal wastewater effluent, brackish groundwater, industrial wastewaters, and seawater are all purified using RO membranes to produce ultra-pure water for potable, agricultural, and industrial purposes [9]. Engineered osmosis processes, such as FO and pressure retarded osmosis (PRO), may further diversify our water future through low-energy desalination, membrane bioreactors, salinity gradient power, and osmotic energy recovery [10–12]. Membranes can be formed with a wide range of structures, chemistries, and performance characteristics by changing the material or altering the fabrication process [7]. The possibilities are seemingly endless, but effective material tailoring requires some degree of *a priori* knowledge. This intuition is difficult to gain due to the lack of predictive models relating membrane structure and chemistry to performance and a less efficient trial-and-error approach is often taken.

A fundamental understanding of the mechanisms at play in membrane separations is crucial to furthering the field and intelligently tuning materials to meet the presently growing needs and applications [13]. Transport models are the tools used to understand membrane transport. Two primary categories exist: phenomenological and mechanistic models [14]. Phenomenological models treat the membrane as a “black box” and describe the separation process in terms of measureable parameters, such as solute passage and water flux. Empirical coefficients are used to represent the separation performance of a membrane by relating measured concentrations in the feed and the permeate streams. While these can be used to characterize performance they offer no mechanistic explanation of separation and are, therefore, difficult to compare between materials. Mechanistic transport models have been developed to relate the separation performance observed in a membrane to the physical (*e.g.*, pore structure, solute size) and chemical properties (*e.g.*, energy of interaction) of the membrane and solute materials. Such structure–performance relationships allow for a more fundamental understanding of membrane transport. Herein, we review the types of membranes and relevant mechanistic transport models for osmotic membrane separations. In each model, we lay out the parameters and methods needed to effectively apply the models, paying particular attention to the inherent advantages and limitations of each.

2. State of the art osmotic membranes

2.1. Integrally-skinned asymmetric membranes

The majority of membranes currently employed in industry are asymmetric porous membranes [15–17]. In asymmetric porous membranes structure and transport properties vary across the membrane thickness. An asymmetric membrane normally consists of a 0.1–1 μm thick dense layer supported by a highly porous, 100–200 μm thick support layer [15,16]. The dense layer provides the majority of selectivity for the membrane. Separation properties are determined by the chemical nature, size of pores (0.4–1 nm), and thickness of the skin layer. The porous substructure is assumed to provide mechanical support for the thin and fragile selective layer and thought to have little effect on the separation performance of the membrane. Recently, however, the effects of the support layer chemical properties (*e.g.*, hydrophilicity/hydrophobicity) and pore structure (*i.e.*, pore size and porosity) on composite membrane transport have been re-evaluated [18–21].

2.2. Thin film composite membranes

In thin film composite membranes, the porous support layer is generally an integrally skinned membrane formed through the non-solvent induced phase separation (NIPS) process. The skin layer is typically formed through either interfacial polymerization or dip coating followed by cross-linking [16,22]. The most common thin film chemistry for RO membranes is based on a fully aromatic polyamide (PA) formed by interfacial polymerization of *meta*-phenylenediamine (MDP) and trimesoyl chloride (TMC), while popular polyamide NF membranes are formed by interfacial polymerization of piperazine and TMC [22]. It is assumed that the dense selective layer formed by interfacial polymerization is heterogeneous throughout its thickness (20–200 nm) and highly cross-linked. The surface properties of a PA film are different from the properties within the PA dense layer because the polymer density is not uniformly distributed [23]. The PA dense layer is highly negatively charged since acyl chloride groups are not fully converted to amide in the formation process; however, it was recently reported that direct titration experiments revealed the simultaneous presence of both positive and negative fixed charges in the dense layer of composite PA NF membranes [24]. Freger and Srebnik proposed that fixed charge is not uniform and that the film is actually a “sandwich” comprising two oppositely charged layers [25].

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