Membrane Fouling

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Membrane-based separation has gained increased popularity over the past few decades, particularly reverse osmosis (RO). A major impediment to the improved performance of membrane separation processes, in general, is membrane fouling. Fouling has detrimental effects on the membrane's performance and integrity, as the deposition and accumulation of foulants on its surface and/or within its pores leads to a decline in the permeate flux, deterioration of selectivity, and permeability, as well as a significantly reduced lifespan. Several factors influence the fouling-propensity of a membrane, such as surface morphology, roughness, hydrophobicity, and material of fabrication. Generally, fouling can be categorized into particulate, organic, inorganic, and biofouling. Efficient prediction techniques and diagnostics are integral for strategizing control, management, and mitigation interventions to minimize the damage of fouling occurrences in the membranes. To improve the antifouling characteristics of RO membranes, surface enhancements by different chemical and physical means have been extensively sought after. Moreover, research efforts have been directed towards synthesizing membranes using novel materials that would improve their antifouling performance.

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1. Introduction

Water desalination is the process of purifying seawater or brackish water from salts and contaminants to produce water suitable for domestic and industrial applications. Membrane-based technologies are a promising approach to water treatment, due to their high energy efficiency, compactness and low space requirement, operational simplicity, and ease of automation. Among the most well-established membrane-based techniques for desalination are, reverse osmosis (RO), microfiltration (MF), nanofiltration (NF), ultrafiltration (UF), and membrane distillation (MD) [1][2].

Fouling is generally defined as the deposition and accumulation of undesired materials on the surface of or inside a given host solid material such as membranes, heat exchangers, and boilers. The deposited materials could be dissolved particles, partially soluble organic and/or inorganic macromolecules and/or biological micro-organisms. Thus, in membrane-based water treatment processes, membrane fouling is an inevitable occurrence that can significantly impair the processes' performance, operation, sustainability, and economic feasibility. Fouling mechanisms are a product of the complex physical and chemical interactions between various feed constituents and the membrane surface [3][4][5]. The major contributing factors to membrane fouling include [6]:

- Feed chemistry and composition, i.e., pH, ionic strength, and foulant concentration.

- Concentration polarization (CP): CP can be broadly described as the deposition of rejected solutes on the membrane's surface, creating a region near the membrane with spatially varying concentrations known as the polarized layer. This added resistance causes an increase in the osmotic pressure across the membrane, which decreases the driving force of the process (transmembrane pressure (TMP)), the permeate flux and the observed solute rejection, all of which increase the possibility of membrane fouling [5][7].

- Membrane properties include membrane material type, porosity, hydrophobicity, surface charges, membrane morphology, and molecular weight cut-off (MWCO).

- Process operating conditions such as temperature, pressure, aeration, permeate flux, and several other hydrodynamic conditions.

2. Membrane Foulants

Membrane fouling is generally classified according to the type of foulant or location of fouling. The main categories of foulants include particulate, organic, inorganic, and biological micro-organisms (biofoulants). As for location, fouling can occur internally or on the membrane's surface. Most low-pressure membrane-based separation processes, such as UF and MF, suffer from internal fouling due to the adsorption and clogging of the pores. On the other hand, relatively denser and more compact semi-permeable membranes, such as those used in NF and RO, experience surface fouling. The following sections elaborate on membrane fouling based on the type of foulant.

2.1. Particulate Fouling

Colloids or colloidal dispersions are solutions in which particles of one substance, usually ranging in size from a few nanometers to micrometers, are evenly dispersed in another substance [8]. Colloids can be classified according to size into settleable colloids where the particle size exceeds 100 µm, supra-colloids where the particle size ranges from 1 to 100 µm, colloidal solids where the particles range in size from 0.001 to 1 µm, and dissolved solids which are less than 0.001 µm in size [9]. Moreover, aquatic colloids can be classified according to the dispersed organic and inorganic compounds. Organic colloids comprise proteins, fats, carbohydrates, greases, oils, surfactants, and bio-colloids; whereas, inorganic colloids include clay, silt, crystals, silica sediments, as well as aluminum and iron precipitates resulting from incomplete treatments [9][10].

Particulate fouling is a multi-stage mechanism; the first stage starts with pore-blocking, during which particles deposit near the pore opening, constricting the aperture. With time, layers of the deposited particles will build up on the initial ply, causing the complete sealing and blockage of the membrane's pores. The second stage is characterized by increased deposition, which forms a thick 'cake layer', compromising the membrane's role in the effective and selective removal of the contaminants. The cake layer formed induces higher hydraulic resistance (referred to as cake resistance) and causes strong CP, which, as mentioned earlier, could decrease permeate flux and increase the adverse effects of the foulants [5][6]. The occurrence of particulate fouling is a function of the membrane surface properties (e.g., morphology and topography), feedwater characteristics, including the types of fouling agents present, their concentrations, and their physicochemical properties such as size and surface charge, the feed's chemistry in terms of solution pH, ionic strength and charge interactions, as well as the operating conditions (i.e., temperature, pressure, flux and cross-flow velocity (CFV)). Generally, smoother membranes with enhanced hydrophilic characteristics and low charge surfaces are less likely to experience particulate fouling, whereas hydrodynamically stressful operating conditions featuring high membrane flux rates and/or low CFV can cause severe membrane damage [11].

2.2. Organic Fouling

Organic fouling is caused by the accumulation and deposition of relatively dense organic materials, such as polysaccharides, proteins, humic substances, nucleic acids, lipids, and amino acids. Dissolved organic matter (DOM) is abundant in both surface water and wastewater and can be classified into natural organic matter (NOM), synthetic compounds, and soluble microbial products (SMPs). NOM is naturally occurring heterogeneous mixtures formed as a result of the decomposition of animal and plant remains. Synthetic compounds are man-made DOMs and are artificially added or generated during the disinfection process. Lastly, SMPs are byproducts of biological treatment processes where organic compounds are biologically decomposed. Relevant research studies suggest that the effect of organic matter on RO membrane fouling varies according to the dominating type of organic foulant, feedwater chemistry, foulant-surface, and foulant–foulant interactions [5][6].

2.3. Inorganic Fouling

Inorganic fouling, or scaling, is the deposition of inorganic compounds on the membrane surface or inside the membrane pores [5]. The deposits could be either inorganic compounds with low solubility in water or solutes present in large amounts in water. They form supersaturated solutions and eventually precipitate out of the solution and onto the surface of the membrane. Scale formation on membrane surfaces could occur through two mechanisms, i.e., crystallization or particulate fouling. The former mechanism involves the precipitation of ions and their subsequent deposition on the membrane, whereas the latter involves the convective transport of particulates from the bulk of the solution to the membrane surface. In rivers, groundwater, seawater, and municipal wastewater, the main inorganic compounds that contribute to scaling are hydroxides, sulfates, carbonates, calcium, magnesium, iron, ortho-phosphates, silicic acids, and silica [5][6].

2.4. Biofouling

Biofouling refers to the adhesion and accumulation of microorganisms accompanied by biofilm development on the solid host material and can account for up to 40% of the total fouling during reverse osmosis filtration [3][12]. Table 1 provides a list of microorganisms that commonly attack membranes [13]. The biofilm is an assembly of surface-associated organisms enclosed within layers of a polymer-like material referred to as the extracellular polymeric substance (EPS) matrix. The EPS matrix is quite robust and is capable of protecting the enclosed microorganisms from biocides and toxins. The EPS matrix's makeup depends on the environment in which the biofilm develops; commonly matrices contain proteins, glycoproteins, lipoproteins, nucleic acids, lipids, and polysaccharides. Mineral crystals, clay, silt, or corrosion particles can also be part of the composition of the matrix [3][5][14]. Membrane surface roughness and hydrophobicity are the two main characteristics that significantly affect biofilm development. Generally, hydrophobic membranes are more prone to microbial-associated interactions, thus, are more likely to experience biofouling. Similarly, membranes with rough surfaces tend to have larger exposed surface areas compared to smoother membranes, which provide more active sites for microbial growth and adhesion, increasing the chances of biofouling [13].

Table 1. Common microorganisms Identified in biofilms (adapted from [13]).

Biofouling has various effects on the physical components and operation of the membrane processes. These effects are clarified below [13]:

- Flux decline: as with the case of particulate fouling discussed earlier, the biofilm formation increases the resistance and reduces the permeate flux.
- Membrane biodeterioration: damage to the membrane's structure due to acidic byproducts resulting from the microorganism's biological activity.
- Deteriorated salt retention: inhibition of conventional transport mechanisms and increased CP effects across the membrane, due to the accumulation of dissolved salts and ions on the surface.
- Increased differential pressure: this is due to the increased resistance caused by biofilm formation.
- Higher energy requirements: the high-pressure requirements and the decline in permeate flux result in increased energy consumption.
- Frequent chemical cleaning: the cleaning process disrupts the membrane plant operation and shortens membrane life.

3. Membrane Integrity and Fouling Diagnosis

Fouling is a burden on membrane-based treatment plants because it adversely affects the overall performance and efficiency of processes. Thus, conducting proper fouling diagnosis is an essential element in the management and control of fouling. Generally, the first step in RO membrane fouling diagnostics is the visual inspection and collection of samples from the malfunctioning membrane vessels. Visual inspection and sample collection are run in parallel with performance data analysis to determine whether the membrane performance challenges were caused by a loss of membrane integrity or productivity-related issues [15][16][17]. Membrane integrity diagnostics involve two steps [15]:

- A thorough assessment of the malfunctioning membrane's conductivity profiles followed by an evaluation of the extent of deviation from expected performance;

- Examination of the malfunctioning membrane's peripheral matrix and identifying any defective components (i.e., interconnectors, end-seals, spacers, O-rings).

The most common and comprehensive method for identifying the nature and source of membrane integrity and fouling is membrane autopsy. This method involves a series of laboratory tests on membrane sections taken from membranes with compromised performance. The following are the steps involved in membrane autopsy:

- External inspection: the different components that constitute the RO membrane are visually examined to diagnose the damaged zones. The core tubes, fiberglass castings, and anti-telescoping devices (ATDs) are carefully checked for potential impairments, including any obvious accumulation of foulants, crystals, scales, and biofilms [15][18][19];

- Weight evaluation: the weight of the defective RO membranes is registered and compared against the weight of new RO membranes of a similar size. A significant increase in membrane weight is indicative of dense membrane fouling [15][18][19];

- Mechanical integrity tests: several direct and indirect techniques have been developed to evaluate membrane integrity. Direct methods mainly utilize pressure-driven approaches to specify any grooves or channels in the sheets of the membrane, whereas indirect methods assess the overall integrity of the membrane's structure [20];

- Dye test: dye testing is used to test damage to the surface of some membrane materials. Commonly used dyes include Congo Red, Methyl blue, and Rhodamine B. A fairly intense color is seen on damaged surfaces, particularly where the damage permits the access of a rather large dye molecule to the exposed surface on the porous supporting layer. If the membrane is intact, a uniformly colored stain would be observed [15][18];

- Cell test: a cell test is carried out to evaluate the performance of the malfunctioning membrane by comparing it against a new one, namely through a comparison of the differences in salt rejection and flux rates. The test is conducted by extracting an autopsied element from the defective membrane, followed by soaking it in deionized (DI) water to clean it from fouling residue and buildup, then inspecting its performance and comparing it against the standard performance of new membrane elements [15][19][21];

- Thorough analysis of the foulants: characterization techniques like scanning electron microscopy (SEM), energy dispersive X-ray (EDaX), Fujiwara oxidation testing, thermogravimetric analysis, and biological reactivity testing are commonly used to depict the membrane's surface conditions and topography, and distinguish the different types of accumulated foulants [15][18].

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