Pulsed Flows and Patterned Surfaces Mitigate Membrane Fouling

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Contributor: August H. Young, Nico Hotz, Brian T. Hawkins, Zbigniew J. Kabala

Patterned membrane surfaces offer a hydrodynamic approach to mitigating concentration polarization and subsequent surface fouling. However, when subject to steady crossflow conditions, surface patterns promote particle accumulation in the recirculation zones of cavity-like spaces. In comparison, a rapidly pulsed crossflow induces mixing mechanisms (i.e., the deep sweep and the vortex ejection) that disrupt recirculation zones and reduce the rate of particle accumulation.

Keywords: filtration ; concentration polarization ; fouling mitigation

1. Challenges to Membrane Operation

Membrane filtration is a popular tool in treating wastewater for reuse [1][2][3]. Compared to other treatment technologies, membranes require less energy than thermal treatment processes and less time than traditional filtration methods (e.g., sand filtration). Further benefits of membrane technologies are modest footprints, relatively low capital costs, and a demonstrated efficacy in producing pathogen-free outputs. However, in application to complex waste streams with high fouling potential, membranes face significant operational challenges, namely concentration polarization and surface fouling.

Although concentration polarization is a reversible phenomenon (often mitigated via disturbance of the solute layer at the membrane surface [4]), it leads to fouling of the membrane surface when left uncurbed [5][6]. This type of fouling significantly thwarts the wide-scale adoption of nanofiltration [7] and poses similar operational challenges to the implementation of microporous membranes (i.e., microfiltration and ultrafiltration modules) [4]. The overwhelming consensus is that these phenomena pose the most significant hurdles in the application of pressure-driven membrane processes [8][9][10][11][12].

Fouling at the membrane surface is typically diminished by chemical modification of the membrane surface or cleaning procedures. The former is undoubtedly beneficial to the targeting and rejection of certain contaminant species but is cautioned by authors who express concerns about stability, longevity, and effect on membrane performance [10][13][14]. Additionally, the latter is routinely used to restore membrane performance but is also associated with several significant drawbacks. Chemicals commonly used in cleaning procedures can deteriorate the physical properties of the membrane surface, reducing selectivity to solutes in the feed stream [15], especially in the case of polymeric modules [16] and other thin film composites [17]. Membranes can also be damaged by incorrect chemical cleaning procedures [18], and one must consider the negative environmental impacts of chemical use [19]. At the very least, operational downtime should also be considered a drawback to any traditional cleaning method that requires taking the membrane offline [12]. Thus, it is advantageous to minimize the need for and frequency of chemical cleanings.

2. Hydrodynamic Solutions

In order to postpone the need for chemical cleaning agents, it is necessary to mitigate particle aggregation at the membrane surface—the precursor to membrane fouling.

2.1. Manipulation of the Feed Flow

Reducing the severity of concentration polarization requires that the solute layer at the membrane surface be disturbed. Most commonly, this is achieved by operating the membrane in crossflow in the transitional/turbulent flow regime to increase wall shear and induce local turbulence; it is widely recognized that low feed Reynolds numbers result in increased concentration polarization [20][21]. This technique, however, can be cost-prohibitive due to the associated energy demand of feed pumps [10][21]. Other commonplace approaches in the crossflow operational mode include limiting the length of the membrane surface (to inhibit boundary layer growth) and utilizing pulsatile flows, flow reversals, and centrifugal instabilities [22]. Jaffrin [23] provides a thorough overview of these techniques and highlights their ability to reduce surface fouling and improve filtration performance. Recent examples of studies that highlight the benefit of pulsed flow over steady include Kürzl and Kulozik [24], Liu et al. [25], and Wang et al. [26]. In another related example, Echakouri et al. [27] use a periodic feed pressure technique to minimize surface fouling.

Although there are a limited number of investigations into the dynamics of pulsed flow in membrane channels with spacers (e.g., see [24][28][29][30]), there are none that combine the study of pulsed flow with patterned membrane surfaces. While both research endeavors involve the simulation of vortex dynamics, the former focuses on the efficacy of using vortex shedding to scour the length of an otherwise flat membrane surface.

2.2. Manipulation of the Membrane Surface

Patterned membrane surfaces, which induce local turbulence and high surface shear, offer yet another hydrodynamic approach to mitigating concentration polarization and surface fouling. Multiple research groups have demonstrated the ability to create micro- and nano-structures on flat and tubular membrane surfaces. Among them, researchers highlight Won et al. [31][32][33], who demonstrated successful fabrication of membrane surfaces with pyramids and prism patterns; researchers replicate the two-dimensional surface pattern utilized by Won et al. [32]. Heinz et al. [34] provide a comprehensive list of thirty-five studies concerning the fabrication of patterned polymeric membrane surfaces used in separation applications. Barambu et al. [35] summarize the approaches used to produce these patterns and the subsequent effect on membrane performance. Chauhan et al. [36], Zare and Kargari [12], and Ibrahim and Hilal [37] provide more recent and nearly concurrent review articles.

Aside from the recent study of Ward et al. [38], there is ample numerical and experimental evidence that patterned membrane surfaces effectively mitigate concentration polarization. Çulfaz et al. appear to be among the first to study the effect of a controlled surface pattern on fouling and membrane performance, excluding previous work on corrugated membranes, as summarized by Ibrahim and Hilal [37]. There was, however, ample interest in the study of spacer patterns in cross-flow channels beforehand (e.g., see Ma and Song [39] for results and a brief summary of prior studies). In their study on the fouling behavior of micro-structured hollow fibers in the filtration of sodium alginate, Çulfaz et al. [11][40] found that, in comparison to smooth fibers, the structured fibers exhibited a higher degree of reversibility in surface fouling. The authors attribute this phenomenon to a looser packing of the deposited particles onto the structured membrane, which is ultimately more conducive to removal procedures. Similar results were obtained by Rickman et al. [41], who found that patterned membranes recover more of their initial flux than their nonpatterned counterparts. Further, Ward et al. [42] found that when exposed to feed streams containing E. coli cells, nanopatterned membranes recovered 18% more of their initial flux than non-patterned membranes. In this case, the improved cleanability of the patterned membrane is attributed to the fact that the E. coli cells cannot deposit into the valleys of the membrane, thus thwarting the rate of biofouling in these regions [42].

Following the work of Çulfaz et al. [11][40], Won et al. [33] utilized a new patterning process to treat wastewater in crossflow; they found that the deposition of microbial cells onto the patterned membrane surface was significantly reduced. The authors attribute the reduction to the apexes of the surface pattern, which they deem responsible for inducing local turbulence and high shear—a conclusion also supported by their later work on the biofouling of prism patterns [31]. Shortly after the work of Won et al. [33], Lee et al. [43] published similar findings, indicating the importance of flow characteristics and shear stress distribution for the frequency and severity of particle deposition in the valleys of the surface pattern. Shang et al. [7] later confirmed that the reduction in concentration and thickness of the concentration polarization layer is attributed to the increased shear generated by the surface pattern.

In another study on triangular-patterned surfaces, Choi et al. [44] highlighted a dependence on the size of the particles in the feed suspension and pore water flux in addition to the crossflow feed rate. The authors found that, for the microfiltration of mixed suspensions (i.e., those with a large distribution in particle size), the deposition of larger particles affected the flow streamlines and therefore the deposition of smaller particles. They also found that the bulk flow and vortex streamlines were “well-separated” from one another, making it difficult for small particles to traverse the separation and deposit onto the valleys of the membrane surface. Malakian and Husson [45] recently used this argument to explain the low levels of protein deposition they observed in the valleys of a herringbone surface pattern. Jung et al. [46] found that the particles they studied tended to deposit into the surface valleys and not at the peaks, indicating a conclusion later drawn by Jung and Ahn [47]: a patterned membrane surface can tremendously reduce surface fouling, given a judicious choice in Reynolds number and pattern depth relative to average particle size. Kim et al. [48] recently offered further confirmation of these dependencies via an alternative approach (i.e., a herringbone-patterned mixer to induce chaotic advection in a flat sheet membrane module).

Won et al. [32] provided a numerical investigation of patterned membrane surfaces, manipulating the parameters of the surface pattern to determine the effect on particle deposition. The authors showed that the tested patterns yielded a significant reduction in the mass attached to the membrane wall. Maruf et al. [10] and Jamshidi Gohari et al. [9] demonstrated a decrease in surface fouling accompanied by an increase in orientation angle between the surface pattern lines and feed flow direction. Malakian et al. [49] highlighted yet another geometric dependence on pattern width, noting the subsequent effect on vortex size and the ability of the vortex to shield the membrane surface from particle deposition (a phenomenon previously termed "vortex-induced shielding" by Choi et al. [50]). Wang et al. [51] provided a thorough summary of the work that has been undertaken, to date, on correlating pattern configuration with the rate of particle deposition and fouling, both numerically and experimentally.

3. A New Combined Approach

Although there is extensive documentation that patterned membrane surfaces reduce the thickness and degree of concentration in the concentration polarization layer, surface patterns produce stationary vortices in valleys and cavity-like spaces capable of trapping buoyant particles. It is recognized that the presence of these stagnant zones promotes particle aggregation and induces surface fouling in the pattern valleys [8][43][46][51][52] and can thus result in an overall higher degree of fouling [12]; the same is true for studies concerning the fouling of membrane channels with spacers. Wang et al. [51] explain that while vortices can create flow separation in pattern valleys and therefore hinder particle deposition along the valley surface, they can also capture foulants. Once captured, foulants generally remain and aggregate in the recirculation zone [52], given the need to traverse sometimes significant flow separations to move back into the bulk flow [32]. For example, in their study of a rectangular surface pattern, Gençal et al. [8] found that an increase in surface pattern height relative to pattern width created larger dead zones in the pattern bottoms and exacerbated surface fouling. Given the tendency of trapped particles to aggregate and aggravate surface fouling in pattern valleys, there remains the need to flush these stagnant zones with a comparatively clean flow volume. This can be accomplished via a rapidly pulsed flow, which, when paired with a cavity-like geometry, induces the deep sweep and vortex ejection mechanisms. Previously, Kahler and Kabala [53][54][55] found that these mechanisms are responsible for enhanced transport between cavity spaces and the bulk flow; they build upon the work of Sobey [56], who determined that enhancement was a result of vortex emptying and filling in cavity-like spaces after Bellhouse et al. [57] designed a pulsatile flow system to create unsteady vortices and enhance gas transport in furrowed channels. In simulation, Kahler and Kabala [53] found that rapidly pulsed pumping in the dead-end pore space recovered 11% more contaminant than steady flow. They also found that rapidly pulsed pumping recovered the same amount of contaminant as steady flow seven times faster. Later, Kahler and Kabala experimentally verified accelerated removal in column testing on granular media [55]. Therefore, it is plausible to suspect that rapidly pulsed pumping could disrupt concentration polarization at the membrane surface via the deep sweep and the vortex ejection mechanisms. Given the ability of these mechanisms to move otherwise trapped particles back into the bulk flow, a rapidly pulsed feed flow could significantly reduce the fouling of patterned membrane surfaces.

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