AN INVESTIGATION OF HYDRODYNAMIC CONDITIONS INSIDE GAS-SPARGED HOLLOW FIBER MEMBRANE MODULES

by

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Abstract

Over the past decade, membrane filtration has emerged as a proven technology for water and wastewater treatment applications. Despite its popularity, the problem of membrane fouling remains the Achilles heel of membrane filtration. A common strategy to control membrane fouling is the use of gas sparging to prevent particle deposition on membrane surfaces. The efficiency of gas sparging for fouling control/prevention depends on the effective distribution of sparged gas bubbles and bubble-induced flow across membrane surfaces. To date, there is very limited literature available that describes this hydrodynamic condition inside the submerged hollow fiber membrane modules. The reason for this limited knowledge is the complex and transient nature of the geometry and hydrodynamics inside hollow fiber modules. The hydrodynamic conditions surrounding a hollow fiber under gas sparging, and the relationship between hydrodynamic conditions and fouling control are not well understood. This presents an obstacle to optimizing the performance of submerged hollow fiber modules with respect to energy costs associated with gas sparging. This thesis provides a comprehensive study of the hydrodynamic conditions inside a gas-sparged submerged hollow fiber membrane module, and the relationship between the observed hydrodynamic conditions and fouling control. Unlike what had been hypothesized by some, the results indicated that the hydrodynamic conditions inside a submerged hollow fiber membrane are different than those of confined tubular membrane systems. It was also observed that different types of shear profiles exist inside the membrane module, and the different types of shear conditions result in different fouling, which suggests that different mechanisms are at play in controlling particle transport near the membrane surface. This information opens the opportunity for further investigation in terms of optimization of the gas -sparging system, or other shear-generating devices that create shear conditions that offer the greatest benefit minimizing fouling, while minimizing the energy demand associated with generating these shear conditions.

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Nomenclature

а	Particle diameter	m
А	Area of electrode	m^2
A _m	Area of membrane (Equation 3-3)	m ²
c	Solute concentration	kg m ⁻³
ci	Concentration of species <i>i</i>	mol m ⁻³
c _b	Bulk concentration	mol m ⁻³
С	Ferricyanide concentration	mol m ⁻³
Co	Bulk concentration	mol m ⁻³
C_1^B	Concentration of reacting species	mol m ⁻³
$C_{salt}^{\ B}$	Concentration of salt	mol m ⁻³
d	Electrode diameter	m
d _{cor}	Corrected electrode diameter	m
D	Diffusion coefficient	$\mathrm{cm}^2\mathrm{s}^{-1}$
D_b	Diameter of bubble (Equation 3-12)	m
Di	Diffusion coefficient of species <i>i</i>	$\mathrm{cm}^2\mathrm{s}^{-1}$
f	Frequency of flow fluctuation or shear event	s^{-1}
f_p	Frequency of pulsing	s^{-1}
f_r	Friction factor for laminar pipe flow	
F	Faraday's constant	96287 C / equiv
g	Gravitational acceleration	m s ⁻²
g _{cor}	Geometric calibration factor for probe	
G	Signal amplifier/gain	
I ₁	Total current	Ampere
I _{mig}	Migration current	Ampere
I _{px}	Measured current of Probe X	Ampere
I _{p1}	Measured current of Probe 1	Ampere

J	Volumetric flux	m s ⁻¹
J _o	Clean membrane volume flux	$m s^{-1}$
$\overline{\mathbf{k}}$	Coefficients index in Hermia's models	units depend on mechanisms
k	Mass transfer coefficient	$m s^{-1}$
k _c	Cake filtration constant	kg m ⁻³
k _B	Boltzmann constant	$1.3806503 \times 10^{\text{-}23} \text{ m}^2 \text{ kg s}^{\text{-}2} \text{ K}^{\text{-}1}$
K _c	Cake formation constant	s m ⁻²
K _b	Complete blocking constant	s ⁻¹
K _i	Intermediate blocking constant	m^{-1}
Ks	Standard blocking constant	m^{-1}
L	characteristic length of bubble	М
Lg	Length of gas slug	М
LL	Length of liquid slug	М
М	Molecular weight	kg mol ⁻¹
n	Coefficients index in Hermia's models,	units depend on mechanisms
	defined by Equation 3.2	
n _p	Number of pores per unit area	m^{-2}
Ni	Flux density of species <i>i</i>	mol cm ⁻² s ⁻¹
Ns	Number of shear event	
Р	Pressure	Pa
Po	Clean membrane filtration pressure	Pa
r _p	Pore radius	М
r _i	Generation of species <i>i</i>	
R _c	Cake resistance in surface transport models	m^{-1}
R	Ohmic resistance	Ω
R _u	Universal gas constant	$8.314 \text{ x } 10^3 \text{ kg m}^2 \text{ s}^{-2} \text{ K}^{-1} \text{ mol}^{-1}$
R _m	Membrane resistance	m ⁻¹
Ro	Clean membrane resistance	m^{-1}
R _T	Total filtration resistance	m ⁻¹
S	Rate of cake erosion per unit area	$kg m^{-2} s^{-1}$
t	Time	S

Т	Temperature	Κ
T _{max}	Duration of peak shear	S
TMP	Transmembrane pressure	Pa
ui	Mobility of species <i>i</i>	$cm^2 mol J^{-1} s^{-1}$
V	Number of ions formed during dissociation	
v	Molar fraction of species	
v	Velocity	m s ⁻¹
Ve	Number of electrons involved in transfer	
v _i	Average velocity of species <i>i</i>	m s ⁻¹
V	Volume filtered (Chapter 3)	$m^3 m^{-2}$
V	Voltage drop (Chapter 4)	V
V _{cal}	Corrected voltage	V
Zi	Charge number of species <i>i</i>	
V _X	Velocity in x direction	$m s^{-1}$
$\mathbf{V}_{\mathbf{y}}$	Velocity in y direction	m s ⁻¹
Vz	Velocity in z direction	$m s^{-1}$
у	Distance y in viscous sublayer	М
α	Cake specific resistance	m kg ⁻¹
β	Gas volume fraction	
δ	Channel width	М
δ_{μ}	Membrane thickness	М
δ_{μ}	Concentration boundary layer thickness	М
Φ	Potential field	
ϕ_{β}	Bulk particle volume fraction	
ϕ_{ω}	Particle volume fraction at the wall	
γο	Shear rate	s ⁻¹
η	Suspension kinematic viscosity	$m^2 s$
μ	Dynamic viscosity	Pa s
$\mu_{ m L}$	Liquid dynamic viscosity	Pa s

π	Osmotic pressure	Pa
ρ	Suspension density	kg m ⁻³
$ ho_g$	Gas density	kg m ⁻³
$ ho_L$	Liquid density	kg m ⁻³
σ	Interfacial tension	$N m^{-1}$
τ	Shear stress	$N m^{-2}$
$\frac{1}{\tau}$	Time averaged shear	Pa
$ au_{amp}$	Amplitude of shear	Pa
$ au_{gas}$	Gas slug shear stress	$N m^{-2}$
$ au_{Liq}$	Liquid slug shear stress	$N m^{-2}$
$ au_{max}$	Maximum shear stress	$N m^{-2}$
$ au_{min}$	Minimum shear stress	$N m^{-2}$
$ au_{\it std}$	Standard deviation of shear profile	Pa
$ au_{tot}$	Total shear stress	$N m^{-2}$
$ au_{\omega}$	Wall shear stress	$N m^{-2}$
${ au_\omega}^*$	Critical erosion wall shear stress	$N m^{-2}$
$\overline{\gamma}$	Mean shear rate	s^{-1}
$\overline{\tau}$	Mean shear stress	$N m^{-2}$
$\overline{\tau_{two-phase}}$	Two-phase time averaged shear stress	$N m^{-2}$
$\overline{\tau_{\sin gle-phase}}$	Single-phase time averaged shear stress	N m ⁻²

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For Raymond and Isaac

1. Introduction

Membrane filtration processes have been used in many biochemical, food and chemical industries for many years. In the last decade, membrane filtration has become more popular in larger scale water and wastewater treatment applications. One of the main reasons for the increasing popularity in the use of membranes over conventional treatment technologies is the emergence of new and lower cost membranes, thus rendering membrane processes more economical. Moreover, compared to conventional technologies, membrane processes also offer many advantages, some of which include: (1) a smaller footprint, (2) lower treated water turbidity, (3) higher volumetric loadings, and (4) reduced sludge production in wastewater treatment.

Despite the many benefits of membrane filtration processes, as well as the decreasing costs associated with membranes, the unavoidable problem of membrane fouling remains associated with membrane filtration processes. Membrane fouling causes a reduction in total permeate yield, increases operational difficulties and ultimately increases the overall operational and capital costs associated with the processes. As such, the problem of fouling is sometimes coined the Archilles heel of membrane filtration processes. In the past few years, there has been considerable amount of effort in attempting to understanding membrane fouling by the scientific community. Between 1994 and 2007, the number of publications dedicated to the topic of fouling in membrane bioreactors increased 4 fold, from around 100 to 400 [1]. However, as Meng et al. [1] pointed out, despite the increased effort, there remains much confusion about fouling.

One thing that is known is that mass transfer at the membrane surface has a substantial impact on fouling. As such, many strategies have been developed to control the mass transfer of foulants to membrane surfaces in order to minimize fouling, and to improve achievable filtration permeate flux. One such popular strategy is the control of hydrodynamic conditions near membrane surfaces to prevent particle deposition on the membranes. Air sparging at the bottom of membrane units is commonly used to achieve

these favorable hydrodynamic conditions. However, to date, the mechanisms of fouling control by air sparging are poorly understood. More specifically, the hydrodynamic conditions inside a membrane module during sparging have not been comprehensively characterized. In addition, the relationship between the hydrodynamic conditions and mass transfer is not understood at a fundamental level. As a result, there is no complete model available to predict the behavior of membrane filtration processes when the operating conditions, as well as the feed water characteristics, are changed. Pilot–scale testing, which is a very expensive and time consuming, is therefore always required to obtain the required design parameters before a full-scale system can be built.

The reasons for the knowledge gap discussed above are two fold: the first is the complicated nature of fouling in water and wastewater systems, and the second is the complicated nature of fluid flow in a membrane module during sparging. This is especially true in a submerged hollow fiber membrane unit in which thousands of fibers are held in close proximity, with bubbles rising between and around the fibers. The knowledge gap therefore, provides the motivation for this thesis, which aims to present a better understanding of the hydrodynamic conditions inside a submerged hollow fiber membrane module under sparging conditions, it also attempts to establish a link between hydrodynamic conditions and fouling control.

In this thesis, the following questions are addressed:

- 1. Do different hydrodynamic conditions exist inside a gas-sparged submerged hollow fiber membrane module during different operating conditions and with different membrane geometries?
- 2. How do the different hydrodynamic conditions affect membrane fouling?

Chapters 4 to 7 are dedicated to answering Question 1, while Chapter 8 is dedicated to answering Question 2. In Chapter 4, the development of an experimental tool capable of quantifying hydrodynamic conditions inside the membrane module is presented. In Chapters 5 and 6, the typical shear profiles inside submerged hollow fiber membrane

surfaces are characterized for different membrane configurations and different operating conditions. In Chapter 7, the distribution of shear profile on hollow fibers in a bundle is examined. In Chapter 8, the relationship between fouling and the different hydrodynamic conditions observed in Chapters 5 to 7 is investigated. A brief introduction to the fundamentals of membrane filtration processes, and a review of relevant literature pertaining to membrane fouling and hydrodynamics are presented in Chapters 2 and 3.

2.1 Membrane Filtration Process

Membrane filtration processes involve the use of a selective barrier to separate the desired species from a solution. Membrane filtration processes commonly used in water and wastewater treatment are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). The principle of the MF and UF processes is based on the sieving mechanism, where the removal of particles and colloidal material is achieved through size exclusion depending on the pore sizes of the membranes. For NF and RO processes, the removal of dissolved constituents (i.e. ionic species) is mainly achieved through a diffusion mechanism, where ionic species are separated from the liquid by the process of diffusion across the membrane.

Membrane filtration processes typically have three process streams – the feed, the permeate and the retentate. The feed is the influent solution to be filtered to the membrane module. The permeate is the filtered effluent from the membrane module. The retentate, also commonly known as the concentrate, is the stream that contains the retained or rejected constituents. The permeate flux is the rate at which permeate passes through the membrane, expressed as a unit volume of permeate per unit area of membrane surface per time. A commonly encountered unit for permeate flux is LMH (liter $/m^2/hour$).

The driving force of filtration in MF, UF, NF and RO is transmembrane pressure (TMP), defined as the difference between the feed side pressure and the permeate side pressure:

$$TMP = \Delta P = P_{feed} - P_{permeate}$$
 [2-1]

Pressure on the feed side, or vacuum pressure on the permeate side is typically applied to bring about the desired TMP for filtration. Typical operating pressures, pore sizes, and components filtered for each of the four membrane processes that are commonly used in water and wastewater treatment that are presented in Table 2-1.

memorane processes	· [4]				
Membrane Process	Pressure	Pore Size	Pathogens	Organics	Inorganics
	(bar)	(nm)	removed	removed	removed
Reverse Osmosis	10 - 30	0.1 – 1	C, B, V	DBPs, SOCs	All
Nanofiltration	4 - 20	1 - 10	C, B, V	DBPs, SOCs	Divalent, some
					monovalent
Ultrafiltration	0.1 - 2	10 - 100	C, B, V	None	None
Microfitration	0.1 – 2	100 - 1000	С, В	None	None

Table 2-1. Typical pore size, operating pressure, and components filtered for the four membrane processes [2]

Pathogens: C=cysts. B=bacteria, V=viruses

Organics: DBPs = disinfection by-products, SOCs = Synthetic Organic Compounds

2.2 Membrane Performance

For pressure driven filtration processes, the permeate flux obtained during filtration can be described by Darcy's Law [3]:

$$J = \frac{\Delta P - \Delta \pi}{\eta R_T}$$
[2-2]

For a dilute solution, osmotic pressure of the retained material can be calculated using the van't Hoff equation [3]:

$$\pi = \frac{vcRT}{M}$$
[2-3]

Depending on the components on the feed side, the osmotic pressure $(\Delta \pi)$ may be negligible if the molecular weight of the solute being retained is high. This usually applies to microfiltration and ultrafiltration processes where the components on the feed side consist of colloidal particles.

The total membrane resistance is a sum of resistance due to the membrane itself, as well as that due to fouling. Membrane fouling is defined by the International Union of Pure and Applied Chemistry (IUPAC) as a "process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores" [4]. Fouling increases the resistance to permeate flow, which results in a decrease in permeate flux under constant applied pressure conditions, or an increase in applied pressure under constant permeate flux conditions. Over time, when fouling cause an increase in resistance beyond a maximum acceptable level, the membrane modules are taken offline for cleaning to remove the foulants from the membrane [2]. Membrane cleaning can be achieved physically (e.g. backwash) or chemically.

Resistance due to fouling is dependent on process conditions, which is discussed in more detail in Chapter 3. Resistance due to the membrane itself is independent of process conditions. Instead, it is related to intrinsic membrane properties such as membrane thickness, nominal pore size, membrane tortuosity, porosity and pore size distribution [3]. For an idealized membrane pore which is assumed to be a cylindrical capillary with uniform radius, membrane resistance can be calculated based on Poiseuille flow as [3]:

$$R_m = \frac{8\delta_m}{n_p \pi r_p^4}$$
 [2-4]

In most cases, membrane pores are interwined and tortuous instead of being a straight cylindrical channels, and as such, a tortuosity factor may be applied to Equation 2.4.

In wastewater treatment applications, activated sludge has been characterized to exhibit non-Newtonian behavior [5]. For water applications, permeate viscosity is typically the same as that of water viscosity, and can be calculated as:

$$\eta_p = \frac{479 \cdot 10^{-3}}{(T+42.5)^{1.5}}$$
 [2-5]

2.3 Membrane Material

Membrane material used for water/wastewater applications can be made from organic material or inorganic material. Organic membranes are typically made of synthetic polymers such as polyvinylidene difluoride (PVDF), polyethylsulfone (PES), polyethylene (PE) and polypropylene (PP) [6]. Inorganic matericals such ceramic and metallic membranes have been used, although they are not as common as the polymeric membranes. One reason is the relative cost of the inorganic membrane, which may be ten times higher [7]. However, inorganic membranes do offer some advantages over organic membranes, since they can typically tolerate higher temperature, higher pressure and harsher chemical conditions during membrane cleaning. Different materials used for the membrane also have different surface characteristics such as hydrophobicity and surface charge. The applicability of each membrane material in a membrane filtration process depends on the characteristics of the water matrix as well as the operating conditions.

The structure of the membrane can be either isotropic or anisotropic. Isotropic membrane structures have a uniform membrane composition/structure, while anisotropic membranes consist of different layers with different structure and composition. For example, a typical hollow fiber membrane generally consists of a thick porous support with a thickness of approximately 100 μ m, and a thin membrane skin with a thickness of 0.2 to 0.25 μ m [8].

2.4 Membrane Configuration

The different membrane configurations used in water and wastewater treatment applications can be divided into two classes based on their geometry –tubular and planar. Tubular membranes with diameters greater than 5 mm are known as tubular membranes, while diameters in the range of 0.5 to 5 mm are known as capillary membranes or hollow fibers. Tubular membrane modules can be designed with one single tube placed in a

casing/cartridge, or they can be packed together in a bundle inside a casing/cartridge. Pressurized feed is delivered inside the tube and permeate flow is collected outside the tube. Hollow fiber membrane modules consist of many individual fibers packed in a bundle and potted at one or both ends. Feed flow can be delivered outside the fiber (outside-in), with the permeate collected inside the lumen of the fiber (inside-out), or vice versa. For example, feed flow in the submerged hollow fiber modules used in membrane bioreactors is typically outside-in.

Planar membranes include plate and frame membranes and spiral wound membranes. Plate and frame membrane can be either pressure sealed, or not. Pressure sealed plate and frame membrane modules consist of support plates and sheets of membranes. Two membrane sheets, separated by a spacer, are sandwiched between the support plates, and the entire module is pressurized. Permeate flow is induced due to the high pressure in the feed side, and permeate is collected from the membrane elements.

More commonly, in MF and UF applications, such as a membrane bioreactor, nonpressurized plate and frame membranes are used. This provides additional cost savings since the process can be operated in relatively low TMP, and no frame is required. In this configuration, two membrane sheets are placed parallel to each other, and sealed along the edges. Spacers are placed between the sealed membranes to prevent membranes from touching each other, and placed in a cassette, which may house as many as 150 sealed membranes. The cassettes are immersed directly into a feed tank which is open to the atmosphere. Suction is applied to the inside of the sealed sheets and the flow is outside-in. This membrane configuration is also often referred to as a flatsheet membrane. An example of this type of membrane is the Kubota flat sheet membrane.

Spiral wound membranes consist of two membrane sheets and a spacer sandwiched together and wound spirally along around a central tube. The wound leaf is separated by feed-side spacers. Three edges of the sheets are glued together, and the remaining open edge is connected to a tube where the permeate is collected.

In water and wastewater applications, where MF and UF membranes are typically used, tubular, hollow fiber and flat sheet membrane configurations are most commonly used. Spiral wound and plate and frame membranes are normally associated with nanofiltration and reverse osmosis. Typical operating conditions for these different membrane technologies are shown in Table 2-2. It is noted that the energy consumption rates presented in Table 2-2 are decreasing rapidly due to advances in module design. Currently, a typically accepted energy consumption rate in a MBR for wastewater treatment is around 1 kWh/m³.

Membranes can be operated in dead-end or crossflow mode, shown in Figure 2-1. In dead-end mode, all materials in the feed side are transported towards the membrane via permeate drag, and accumulate on the membrane surface. In the cross-flow mode, a tangential flow is applied across the membrane such that some material is transported away from the membrane surface. Operating in cross-flow mode reduces the extent of membrane fouling and reduces the TMP required to achieve a targeted permeate flux. However, operation in crossflow mode can result in higher energy consumption. In systems where the solid content in the feed is low, a semi dead-end mode is sometimes employed during operation. Semi dead-end systems are operated like dead-end mode but with intermittent membrane cleaning during operation. Membrane cleaning can be chemical or physical, such as backwashing or backpulsing. The frequency of cleaning depends on the feed matrix, the membrane characteristics as well as the operating conditions.

Membrane	Operating	Rate of Flux	Energy Consumption*
Technology	TMP (kPa)	$(L/m^2 d)$	(kWh per m')
microfiltration	7-100	405-1600	0.4 (TMP 100 kPa)
ultrafiltration	70-700	405-815	3.0 (TMP 525 kPa)
nanofiltration	500-1000	200-815	5.3 (TMP 875 kPa)
reverse osmosis	850-7000	320-490	10.2 (TMP 1575 kPa)

Table 2-2. Typical operating conditions of the different membrane technologies [2]

*Energy consumption for the entire membrane system



Figure 2-1. Dead-end vs. crossflow mode

2.5 Hollow Fiber vs Flat Sheet Membrane

Both submerged hollow fibers and flat sheet membrane modules are popular in membrane bioreactors. However, compared to flat sheet modules, the hollow-fiber modules have a higher specific membrane area, are generally less expensive to manufacture, and can tolerate vigorous backwashing [9]. However, due to the high packing density of most hollow fiber modules, even bubble distribution within the hollow fiber bundle is difficult to achieve. This results in uneven fouling control within the module. Bubble distribution, or control of hydrodynamics inside a flat sheet membrane may be easier to achieve. High pressure drop inside the hollow fiber is also another disadvantage of hollow fiber membranes, compared to flat sheet membranes.

2.6 Hollow Fiber Membranes

Hollow fibers are relatively long and narrow, with fiber length in the range of 1 to 2 m and fiber diameter between 0.5 to 2 mm [8]. The fibers are bundled together and both ends are potted in a resin such as epoxy. A typical hollow fiber module may consist of anywhere from a few hundred to 10,000 fibers [8]. Hollow fibers may be operated in an inside-out or outside-in configuration, as shown in Figure 2-2. In the inside-out configuration feed water enters the lumen of the fiber and filters through the skin of the membrane. In the outside-in configuration the feed water is outside of the fiber lumen and the permeate is collected inside the fiber lumen. Inside-out configuration is susceptible to the clogging of the lumen, while offering a lower filtration surface area. Additionally, there is more head loss in the inside-out configuration, while only the permeate flows through the lumen in the inside-out configuration, while only the permeate flows through the lumen in the outside-in configuration. Fibers in the outside-in configuration for fibers in the outside-in configuration. Fibers in the seen found to result in better fouling control [10].



Figure 2-2. The inside-out and outside-in hollow fiber configurations

Depending on the application, operation of hollow fiber modules is pressure-driven or vacuum-driven. Hollow fibers are generally in an inside-out configuration for pressure driven operation, and in outside-in configuration for а vacuum-driven operation. In pressure-driven processes the driving pressure ranges from 20 to 280 kPa, where the pressurized feed is delivered to the membrane module via an external pump [8]. In some cases the external pressure is aided via gravity flow, depending on the layout of the treatment plant. For vacuum-driven processes, the membrane modules are submerged in a tank of feed water, and the lumens in the module are connected to a vacuum pump. Typical suction pressure in a vacuum-driven operation ranges from -20 to -80 kPa [8]. Recently, there have been new additions of pressuredriven outside-in modules for water treatement, where the feed container is pressurized to provide the driving force for permeate flow across the membrane (i.e. ZW2000 systems).

3. Literature Review -Fouling and Hydrodynamics

During the process of water or wastewater filtration, membrane fouling causes an increase in the resistance to permeate flow, which manifests itself as either an increase in the transmembrane pressure (TMP), or a decrease in permeate flux over time. The problem of membrane fouling affects the overall performance of the membrane process and becomes a costly aspect of membrane filtration.

Fouling of membrane surfaces may appear in three general forms: (1) the accumulation of material found in feedwater, (2) the formation of chemical precipitates due to the chemistry of feedwater and (3) the modification and damage of the membrane surface due to biological agents that colonize the membrane surface, as well as chemical substances that exist in the feed water [2]. In this chapter, the focus of the review of literature will be primarily on fouling due to the accumulation of particles at membrane surfaces. The accumulation of particles at membrane surfaces results in the formation of a "cake layer", sometimes referred to as cake fouling. Factors that affect this type of fouling, and the mitigation of membrane fouling through the control hydrodynamic conditions inside submerged hollow fiber membrane modules will be identified as well.

3.1 Types of Fouling

Different types of fouling can occur on a membrane surface. In the literature, the terms "reversible fouling" and "irreversible fouling" are commonly encountered. Reversible fouling is encountered when the loss of filterability is temporary and reversible. Reversible fouling can be defined as hydraulically reversible and chemically reversible.

Hydraulically reversible fouling can generally be described as the build-up of a cake layer, consisting of solid particles and colloids. Recovery of membrane filterability in this case can be achieved through physical membrane cleaning such as backwashing. Chemically reversible fouling can be described as the adsorption of macromolecules onto membrane pores, thus contributing to the narrowing of the pore channels. Recovery of membrane filterability can be achieved through chemical cleaning to remove the adsorped macromolecules from the pores.

Irreversible fouling is normally described as a loss of permeability (or loss of membrane performance) that is permanent and not reversible, even after membrane cleaning (physical and chemical). The main culprits in irreversible fouling are the formation of chemical precipitates, and the permanent damage to the membrane surface due to the biological or chemical agents, which may result in permanent blockage of pores and changes to the membrane's physico/chemical characteristics. Irreversible fouling is generally not affected by the hydrodynamic conditions near the membrane surface. However, the extent of irreversible fouling is relative and is dependent on the frequency of backwashing, as well as the strength of adhesion of the clogging particles from pores and precipitates from membrane surfaces [11].

There are conflicting observations in the literature with regards to the type of fouling that dominates in terms of increased resistance during the filtration of wastewater. Choo and Lee [12], Defrance et al. [13], Bae et al. [14] and Lee et al. [15] found that reversible fouling resistance due to cake formation dominates during the filtration of sewage. On the other hand, Bouhabila et al. [16] and Hwang et al. [17] found irreversible fouling due to membrane clogging by the colloidal fractions dominates. These contradicting studies are likely due to the different operating conditions used during the experiments, the biological states of the suspensions, as well as the configurations of the membrane [17, 18]. In addition, the contradictions are also related to the fact that many studies define irreversible fouling at that which cannot be removed hydraulically. In general, however, the extent of irreversible fouling is more impacted by electrokinetic effects such as membrane material (e.g. membrane charge and hydrophobicity), bulk organic

concentration and solution conditions (e.g. pH and ionic strength) than the hydrodynamic conditions imposed near membrane surfaces [19]. On the other hand, the extent of reversible fouling, particularly cake formation on membrane surfaces is very much dependent on the hydrodynamic conditions near membrane surfaces. The focus of the remainder of the discussion in this chapter will be on reversible fouling. Several parameters that affect the formation of a reversible cake layer are discussed in the subsequent section below.

3.2 Factors that Influence Reversible Fouling

Particle properties (i.e. size, shape, electrokinetic interactions), membrane properties (i.e. pore size, hydrophobicity), operating conditions (i.e. flux, transmembrane pressure, hydrodynamics conditions), and feed characteristics all affect the extent of reversible membrane fouling. The sections below discuss briefly the contribution of these factors to reversible fouling. The hydrodynamic condition near a membrane surface has a substantial impact on controlling fouling. A separate discussion (in Section 3.4) is dedicated to the effect of gas sparging on hydrodynamic conditions near a membrane surface.

3.2.1 Effect of Particle Size on the Cake Structure

In most membrane applications, the particles in the feed stream are not monodispersed. Rather, there exists a range of particle sizes in suspension [2]. The fouling propensity of monodispersed particles is likely different from that of polydispersed particles under the influence of crossflow hydrodynamic conditions. For monodispersed suspensions, Lee and Clark [20] found that hydrodynamics had no effect on the resistance due to the cake layer cake during the filtration of their monodispersed particles in a stirred cell experiment. In contrast, for polydispesed particles, crossflow conditions were observed to have an impact on the type of cake layer formed during filtration, and cake resistivity. Many researchers observed that in a feed matrix consisting of polydispersed particles, the cake layer formed at membrane surfaces consisted of the finer particles among all those particles present in the bulk fluid during crossflow filtration [18, 21-26]. This phenomenon can be attributed to the effect of particle size on the different particle back-transport mechanisms. For particle sizes greater than 1 μ m, back-transport mechanisms such as shear-induced diffusion or inertial lift are effective in transporting particles away from surfaces. The extent of the particle back-transport is dependent on the shear stresses near the membrane surface. For particles of less than 0.1 μ m, the dominant back-transport mechanism is molecular diffusion, which is independent of shear stresses near the membrane surface [3]. Therefore, in polydispersed suspensions which consist of both fine particles smaller than 1 μ m, and larger particles greater than 1 μ m, regardless of the imposed shear near the membrane surface, smaller particles will be transported towards the membrane surface due to the convective permeate flow, while larger particles are more effectively transported away due to the imposed shear flows.

3.2.2 Effect of Particle Shape on Cake Structure

The resistances to flow through a cake layer (e.g. specific cake resistivity) can be affected by the shape of the particles in the cake layer. Since the surface area and volume of the particle is influenced by the particle shape, different particle surface areas and volumes result in different specific areas available for flow. This therefore, changes the porosity of the cake layer [24]. As a result, the resistance to flow through the cake layer during filtration is affected. Wakeman [27] showed that for different particle shapes (cubic, rectangular, spherical, fibrous, cylindrical and flakey), the calculated theoretical specific cake resistance can be quite variable. This is summarized in Table 3-1.

Table 5 1. Influence of particle shape on eake specific resistance [22]		
Particle Shape	Theoretical Cake Specific Resistance	
Fibrous	$1.6 \ge 10^9$	
Cylinder	$1.7x \ 10^9$	
Rectangular	$1.8 \ge 10^9$	
Sphere	3.6×10^9	
Cube	3.6×10^9	
Flake	$1100 \ge 10^9$	

Table 3-1. Influence of particle shape on cake specific resistance [22]

(Specific resistance calculation based on particle size of 10 μ m, porosity of 0.5 and particle density of 2000 kg/m³)

3.2.3 Effect of Particle Electrostatic Interactions on Cake Structure

When particles are compacted together in a cake layer, the electrostatic interactions between the particles can play an important role in determining the porosity of the cake structure. The interaction force between particles is dependent on the balance between the attractive force (e.g. van der Waals forces) and the electrostatic repulsive forces. This balance is a function of the separation distance between the particles. If the repulsive force dominates over the attractive force, then the net particle interactive force will result in a higher porosity in the cake structure.

In general, very small particles tend to exhibit stronger electrostatic repulsion compared to larger particles. This repulsive force decreases with increasing particle size, and in the range of these small particle sizes, the porosity of the cake decreases with increasing particle size. This can lead to an increase in the cake resistance with increasing particle size. For larger particles, there exists a critical particle size where the electrostatic repulsive force between particles becomes negligible. In this range of larger particles, the trend is reversed, where increasing particle size now results in increased cake porosity and decreased resistance to filtration [18, 20, 28, 29]. Therefore, depending on the size and the charge of the particle, electrostatic forces may become the dominant mechanism of fouling instead of the particle transport mechanisms [29].

Additionally, since the electrostatic force between particles is impacted by solution chemistry, properties such as solution ionic strength and pH play an important role in affecting the structure of the cake layer [30-35]. There have also been several reports of "cake structure collapse" during filtration [31, 36, 37]. This collapse occurs when the inter-particle repulsive forces in the cake layer is exceeded by the pressure increase in the cake structure during filtration. The critical pressure that induces cake collapse is also highly dependent on solution properties such as pH and ionic strength [31]. Typical water and wastewater matrix often consist of particles with varying sizes as well as solution chemistry. Therefore the electrostatic interactions between the particles can vary substantially from one water/wastewater matrix to another. This adds an additional layer of complexity in deciphering fouling studies found in the literature.
3.2.4 Effect of Membrane Properties

If the membrane pore size is larger than the size of the particle in the feed solution, then fouling mechanisms such as pore blocking or pore narrowing may occur during filtration. As such, it is expected that larger pore membranes used in MF will typically result in higher fouling propensity compared to smaller pore membranes used in UF [18]. Also, the type of fouling seen in MF experiments is typically expected to be more irreversible, since the dominant mechanism of fouling can be pore narrowing. On the other hand, the type of fouling seen in UF experiments is more reversible, since the dominant mechanism of fouling in this case is typically cake formation. Although there are numerous studies on the effect of membrane pore size on the extent of fouling, conflicting observations are reported that describe this relationship [38-41]. Part of the reason for the conflicting observations is due to the fact that the different studies were conducted with different feed solution characteristics and operating conditions. Different particle size distributions in the feed solution, as well as hydrodynamic conditions and modes of operation (e.g. constant flux vs. constant pressure) all have an impact on the relationship between membrane pore size and fouling [18].

Membrane hydrophobicity also plays an important role in influencing the extent of fouling. In wastewater treatment, hydrophilic membranes have been found to exhibit lower fouling rates during filtration due to the hydrophobic nature of biomass in the feed suspension[11]. (Hydrophobic biomass has a lesser tendency to attach to the hydrophilic membrane, whose surface is covered with a layer of water.)

3.2.5 Effect of Permeate Flux and Transmembrane Pressure

Operation below a critical flux has been experimentally shown by several researchers to be a viable approach to reduce fouling and thereby reduce costs associated with cleaning and maintenance of the membrane modules [10]. The concept of critical flux, first described by Field *et al.* [42] explains that during the filtration operation "there exists a flux below which a decline of flux with time does not occur" [42]. At this flux, the mass transfer of the retainable suspended solids and solutes *towards* the membrane is

theoretically balanced by the mass transfer of the retainable solids and solutes *away* from the membrane. Although the concept of operating below a critical flux is ideal in controlling fouling, operating at a lower flux means a larger membrane surface area is required to achieve the desired filtered permeate quantity. Moreover, there also have been several reports that indicate that fouling still occurs when operating at fluxes far below the critical flux, although the degree of fouling is not as severe as those when operating above the critical flux [43, 44].

Transmembrane pressure during filtration also plays an important role in the observed rate of fouling on membrane surfaces. While investigating the mechanisms of fouling control during filtration of activated sludge in submerged hollow fiber membrane bioreactors, Hong et al. [45] observed that the rate of permeate flux decline increased for increasing transmembrane suction pressure. The authors attributed this phenomenon to two possible factors: (1) the accumulation of bioparticles on the cake layer due to increased transportation of particles to the membrane as induced by higher permeate flow and (2) the compression of the cake layer due to the drag induced by increased permeate flow, thus increasing the resistance in the cake layer. Beaubien et al. [46] studied the effect of different operating conditions on the performance of an anaerobic membrane They observed two distinct zones with respect to the transmembrane bioreactor. pressure-flux relationship. At low pressures, it was found that the transmembrane pressure has a significant effect on permeate flux. At high pressures however, it was found that hydrodynamic conditions in the reactor governed permeate flux. A possible explanation for this observation is that operating at higher pressure leads to higher flux, which results in a higher convective transport of particles towards the membrane, thus leading to increased cake formation.

Cake resistivity is affected by permeate flux and crossflow conditions. Whether or not a particle is carried to the surface of the membrane to contribute to cake resistance is dependent on two competing forces – the convection drag (i.e. permeate flux) of the particle towards the membrane, and the hydrodynamic force (i.e. crossflow conditions, two phase conditions) which encourage particle back-transport away from the membrane

[3]. If the convection drag force dominates over hydrodynamic forces, the particle will be carried to the membrane surface. If hydrodynamic forces dominate, the particle will not reach the membrane. Cabassud et al. [47] also showed that increasing gas flow rate inside a tubular membrane resulted in the expansion of the cake layer, and a decrease of the cake specific resistance. The expansion of cake is likely the result of shear-induced mechanisms that encourage particle transport away from the membrane surface. It is also noted there that, depending on competition between the permeate flux and the particle back-transport mechanisms, there may be a critical pressure across the cake layer, above which the actively labile cake layer may collapse, forming a more dense deposit [31]. It is noted here that in Cabassud's study, the hydrodynamic condition inside the tubular system is well defined. In a submerged hollow fiber membrane module, however, the hydrodynamic condition is not well understood. As such it is not clear if the mechanism of cake expansion in Cabassud's system also occurs near the hollow fiber membrane surface.

3.2.6 Effect of Particle Concentration

One of the strategies of controlling fouling is to reduce the solids concentrations in the feed. This is often achieved through pre-treatment of the feed prior to filtration. Pretreatment is used to reduce solids content (and for nanofiltraion and reverse osmosis processes, the bacterial count in the feedwater). This may include chemical precipitation of solids in the feedwater, or in cases where a limit on bacterial activity is desired (to reduce bio-fouling), chlorination, ozone or ultraviolet (UV) radiation is employed [2].

There are contrasting reports in the literature with respect to the effect of biosolid particle concentration on flux behavior over time. General convention has it that increasing biomass concentration increases membrane fouling rate and therefore, decreases permeate flux over time. Several researchers have reported that fluxes decrease with increasing particle concentration [48, 49]. However, other researchers have found that particle concentration has no effect on the flux decline within the range of concentrations studied (4 to 8 g/L for Le-Clech et al. [40], 3.6 to 8.4 g/L for Hong et al. [45].) It has

also been reported that there exists a critical concentration, above which the effect of MLSS concentration on fouling was observed (30 to 40 g/L for Yamamoto et al. [50], and 3.6 to 8.4 g/L for Harada et al. [51]). Chang and Fane [52] reported that no clear relationship was observed between flux and MLSS for concentrations of 5 to 15 g/L, but when the concentration was increased from 15 to 20 g/L, the critical flux of the filtration process decreased. A possible explanation for these discrepancies between the studies is that a higher particle (MLSS) concentration results in a higher mass transport towards the membrane. If this mass transport towards the membrane exceeds the capacity of the system to remove the material (i.e. critical flux is exceeded), then higher fouling rate is Additionally, at higher MLSS concentrations, the fluid exhibits a nonexpected. Newtonian behavior, which has an effect on amount of turbulences generated in the systems, and therefore affect the effectiveness of fouling control. Moreover, all these experiments were conducted under different operating conditions (i.e. hydrodynamic conditions and water quality.) Depending on the dominant mechanism of particle transport in the module (i.e. shear induced or inertial lift, or molecular diffusion), the back-transport of particles from membrane surfaces is dependent on bulk concentration, as well as shear rate and particle size [3]. Therefore, it is difficult to compare the results of one experiment to another. However, the contradicting observations in the literature highlight the fact that particle concentration in the module is an important parameter, and its impact on fouling and flux is often coupled with those of other parameters such as particle diameter and hydrodynamic conditions. Moreover, changing MLSS concentration likely impacts biomass characteristics, therefore indirectly influencing fouling [40].

3.2.7 Effect of Feed Matrix Characteristics

Other factors, such as the characteristics of the feed matrix also have an impact on the extent of fouling. The characteristics of the feed matrix, such as that of the activated sludge in a wastewater membrane bioreactor, are complex. In biological activated sludge mixed liquors, the components of the matrix consist of the original substrate found in the feed water as well as cellular material produced during the biological processes, all of

which are variable from process to process and are dependent on the feed water characteristics and operating conditions. Many components of the matrix, such as the suspended particulates, solutes, colloidal fractions and dissolved polymers such as extracellular polymeric substances (EPS), have been found to contribute to membrane fouling in membrane bioreactors. There are varying observations in the literature identifying specific components in mixed liquor as the primary contributors to membrane fouling. Chang and Lee [53] found that the higher the EPS content, the higher the fouling propensity; Geng and Hall [54] found that the soluble EPS was the main contributor to fouling; Wisniewski and Grasmick [55] found that the soluble fractions were the main contributors to fouling; while Defrance et al. [13] and Bouhabila et al. [16] showed that suspended solids and colloidal fractions were the main contributors to membrane fouling, All of the studies above were conducted under different operating respectively. conditions, for different membrane types and configurations, all of which have varying effects on the filtration processes. It is also difficult to differentiate between reversible and irreversible fouling in these cases, since the interaction between the deposited material in the cake layer and the colloidal material in suspension may have the potential of to change the reversibility of fouling [18].

3.3 Modeling of Fouling

In the previous sections, different factors that affect reversible cake fouling were discussed. This section provides an overview of the different studies that attempt to model fouling. Although there is extensive literature on the modeling of fouling phenomena during filtration, these models can generally be classified into four categories [11]: (1) resistance-in-series model, (2) flux decline models, (3) film models and (4) particle transport models.

3.3.1 Resistance-in-Series Model

The resistance-in-series model is one of the simpler types of models for describing the characteristics of membrane fouling, and has been used extensively by many researchers in modeling filtrate flux [41, 42, 48, 56-64]. The resistance-in-series model is based on Darcy's model of resistance during filtration, given as:

$$J = \frac{\Delta P}{\eta R_T}$$
[3-1]

The total resistance is normally defined as the sum of the resistances due to reversible fouling, irreversible fouling, and the intrinsic membrane resistance. Fouling resistance can be calculated using results from a series of filtration experiments, the first of which involves filtration of pure water followed by the filtration of the feed solution. The fouling resistances can then be obtained by subtracting membrane resistance from the total resistance [63]. To quantify the effects of irreversible fouling, an additional filtration-experiment can be performed in which pure water is filtered using membranes from which the cake layer (reversible fouling) has been removed via membrane washing (hydraul and/or chemical). Resistances due to reversible, irreversible fouling and membrane resistance can then be obtained. As pointed out by Chang et al., [52] this approach assumes a complete decoupling of the reversible and irreversible fouling, and this is often difficult to assess experimentally. Although there have been several

publications of resistance values [13, 65-71] in a membrane bioreactor in the literature, the values are highly dependent on system configuration, hydrodynamic conditions and the filtrate quality. Therefore, currently bench scale experiments are required to quantify the effect of each parameter on the total resistance values.

3.3.2 Flux Decline Models

Mechanisms of membrane fouling can be described based on four classical models first proposed by Hermans and Bredee [72]. The fouling mechanisms considered in these models include fouling by particles that are larger than the pore size of the membrane, fouling by particles that are smaller than the pore size of the membrane, and fouling by particles that are approximately the size of the pores. The four models that describe these mechanisms are 1) the cake formation model, 2) the standard blocking model, 3) the complete blocking model and 4) the intermediate blocking model.

Figure 3-1 shows each type of blocking model schematically.



(1) cake formation





(2) standard blocking



(4) intermediate blocking

Figure 3-1. The four classical models on the mechanisms of membrane fouling

The cake formation model describes an accumulation on the membrane surface of particles that are larger than the pore size of the membrane. This accumulation results in the formation of a cake layer, which provides an additional resistance to filtration.

The complete blocking model describes the fouling of membranes by particles which are approximately the same size as the membrane pores. The deposition of these particles on the membrane surface results in a complete blocking of the membrane. The number of pores available for filtration decreases over time, and therefore filtration resistance increases over time. The intermediate blocking model is similar to the complete blocking law, with an additional consideration that not all particles that deposit on the membrane will always block or plug a pore. Rather, there is a chance that some particles will roll on top of the particles that are already blocking the pores. The likelihood of the particle depositing on a pore and completely blocking the membrane pore is taken into account in this model. The standard blocking model describes the fouling of membranes by particles which are much smaller than the membrane pore size. These small particles deposit/adsorb onto the membrane surface as well as on the inner walls of the membrane The deposition/adsorption of the particles on the membrane wall results in pore. decreased pore diameter, which leads to an additional hydraulic resistance during filtration. This model is sometimes referred to as the pore restriction model.

Hermia [73] first developed a comprehensive set of mathematical equations describing the four classical models during constant pressure filtration. In his equations, the models were based on dead-end filtration, and are presented in the following form [42, 73]:

$$\frac{d^2t}{dV} = \overline{k} \left(\frac{dt}{dV}\right)^n$$
 [3-2]

The model also assumes that particle distribution is homogenous, there are no particle interactions, the particles are incompressible, the membranes pores are straight cylinders of equal diameter, a dead-end operating mode with no back transport, and that only one mechanism occur at a time.

Field et al. [42] modified Hermia's equations such that the volume filtered V is replaced by permeate flux J, as seen in Equation [3-3]:

$$-\frac{1}{A_m^2 J^3} \frac{dJ}{dt} = \overline{k} \left(\frac{1}{A_m J}\right)^n$$
 [3-3]

For the cake formation model, a simple cake erosion term was added to Equation 3-3 to incorporate the effects of hydrodynamics during crossflow, as shown in Equation 3-4 [42].

$$\frac{1}{J^2}\frac{dJ}{dt} = \frac{\alpha k_c J}{J_o R_o} - \frac{\alpha S}{J_o R_o}$$
[3-4]

It has been recognized that the different fouling mechanisms described above may occur simultaneously. Many researchers have derived different mathematical formulations that combine the different mechanisms [74-78]. Recently, Bolton et al. [75] provided a comprehensive derivation of mathematical expressions which combined the four fouling mechanisms, for both constant flux and constant pressure operating modes. These equations are summarized in Table 3-2 and Table 3-3.

 Table 3-2.
 Summary of the five constant pressure combined fouling models by Bolton *et al.* [66]

Combined Mechanisms	Equation		
Cake formation, complete blocking	$V = \frac{J_0}{K_b} \left(1 - \exp\left(\frac{-k_b}{k_c J_o^2} \left(\sqrt{1 + 2k_c J_o^2 t} - 1\right)\right) \right)$		
Cake formation, intermediate blocking	$V = \frac{1}{K_i} \ln \left(1 + \frac{K_i}{K_c J_o} \left(1 + 2K_c J_0^2 t \right)^{1/2} - 1 \right)$		
Complete blocking, standard blocking	$V = \frac{J_o}{K_b} \left(1 - \exp\left(\frac{-2K_b t}{2 + K_s J_o t}\right) \right)$		
Intermediate blocking, standard blocking	$V = \frac{1}{K_i} \ln \left(1 + \frac{-2K_i J_o t}{2 + K_s J_o t} \right)$		
Cake formation, standard blocking	$V = \frac{2}{K_s} \left(\beta \cos\left(\frac{2\pi}{3} - \frac{1}{3} \arccos(\alpha) + \frac{1}{3}\right) \right)$ $\alpha = \frac{8}{27\beta^3} + \frac{4K_s}{3\beta^3 K_c J_o} - \frac{4K_s^2 t}{3\beta^3 K_c} \qquad \beta = \sqrt{\frac{4}{9} + \frac{4K_s}{3K_c J_o} + \frac{2K_s^2 t}{3K_c}}$		

Combined Mechanisms	Equation
Cake formation, complete blocking	$\frac{P}{P_{o}} = \frac{1}{1 - K_{b}t} \left(1 - \frac{K_{c}J_{o}^{2}}{K_{b}} \ln(1 - K_{b}t) \right)$
Cake formation, intermediate blocking	$\frac{P}{P_o} = \exp(K_i J_o t) \left(1 + \frac{K_c J_o}{K_i} \left(\exp(K_i J_o t) - 1\right)\right)$
Complete blocking, standard blocking	$\frac{P}{P_o} = \frac{1}{\left(1 - K_b t\right) \left(1 + \frac{K_s J_o}{2K_b} \ln(1 - K_b t)\right)^2}$
Intermediate blocking, standard blocking	$\frac{P}{P_o} = \frac{\exp(K_i J_o t)}{\left(1 - \frac{K_s}{2K_i} \exp(K_i J_o t) - 1\right)^2}$
Cake formation, standard blocking	$\frac{P}{P_o} = \left(\left(1 - \frac{K_s J_o t}{2} \right)^{-2} + K_c J_0^2 t \right)$

Table 3-3. Summary of the five constant flux combined fouling models by Bolton et al. [66]

3.3.3 Film Theory Model (Concentration Polarization)

In addition to the four classical models described above, another mechanism often encountered in membrane filtration is *concentration polarization*. Concentration polarization is defined by the IUPAC as the "concentration profile that has a higher level of solute nearest to the upstream membrane surface compared with the more-or-less well mixed bulk fluid far from the membrane surface" [4]. There has been much debate in the literature with whether concentration polarization exists in different types of filtration processes [79]. Traditionally, concentration polarization is thought of as a phenomenon often encountered in reverse osmosis processes, where the presence of low molecular weight material such as salts and organic macromolecules near the feed side membrane surfaces result in large osmotic pressure [3, 80] (as discussed in Section 2.2). A large

osmotic pressure therefore provides an additional resistance to filtration by reducing the effective TMP-driving force required to maintain the desired permeate flux. Concentration polarization is therefore often encountered in the filtration of macromolecules, i.e. in nanofiltartion (NF) and reverse osmosis (RO). In most particulate ultrafiltration (UF) and microfiltration (MF) applications, the retained material at the feed side membrane surface typically has a much higher molecular weight such that the osmotic pressure across the membrane is negligible. Although material may still accumulate at the membrane surface, the concentration of the accumulated material reaches a point of "gel precipitation" [81]. This additional resistance is not traditionally considered as due to concentration polarization. However, it is also possible that concentration polarization can occur in UF as a result of cake fouling on the membrane surface, where the cake layer may behave as a nanofilter which can retain the macromolecules. The retained macromolecule then forms a concentration polarization layer on the membrane surface [44, 79]. Song and Elimelech [82] developed a model which identifies the conditions under which either a concentration polarization, or a cake layer will form on the membrane surface. A dimensionless number, referred to as the "critical filtration number", N_f was defined. If N_f was above a certain value – a cake layer will be formed. If Nf was below a certain value, only the concentration polarization layer s formed.

The film theory model describes the effect of concentration polarization on filtration flux. In this model, solutes and particles carried to the membrane surface form a concentration polarization layer, or a *film*. Within this layer there exists a concentration gradient where the particle concentration is highest at the membrane wall, and lowest in the bulk solution. The diffusion of particles away from the membrane is driven by this concentration gradient. Equation 3-5 describes the film theory model mathematically [3]:

$$J = k \ln \left(\frac{\theta_w}{\theta_b}\right)$$
 [3-5]

Under laminar conditions, the one dimensional mass transfer coefficient k can be derived using the Levesque solution, assuming linear shear flows in the thin boundary layer (film), and negligible permeate flux [3]:

$$k = 0.81 \left(\frac{\gamma D^2}{L}\right)^{1/3}$$
 [3-6]

The primary particle transport mechanism in the film theory is molecular diffusion, which describes the diffusion of particles from a region of high concentration to a region of low concentration through kinetic interactions. The Stokes-Einstein equation describes molecular diffusion [3]:

$$D = \frac{k_B T}{6\pi\eta a}$$
[3-7]

In modeling filtration flux, the film theory was consistently found to underestimate the fluxes of micron-sized particles by *several orders of magnitude* [3]. This underestimation was famously dubbed the "flux paradox" by Green and Belfort [83]. As it turns out – the film theory is only capable of modeling flux matrices consisting of particles in the sub-micron ranges, for which molecular diffusion is the primary mechanism of particle transport. The diffusion of larger particles (greater than 1 μ m) that often occurs with many feed matrices cannot be adequately described by molecular diffusion in the film theory. The next few models discussed below were then developed to describe the particle back-transport of larger particles.

3.3.4 Particle Transport Models

One of the strategies of fouling control is the reduction of solids near membrane surfaces by promoting backtransport of particles away from the membrane. There have been extensive reports and research on the mechanisms of particle transport at membrane surfaces, ever since the importance of the effect of convective flow in mass transport phenomena was recognized. Numerous models have been developed to describe suspension particle transport near the membrane surface. These models are based on the steady-state balance between the convection permeate drag towards the membrane surface, and the back-transport mechanisms which carry the particles away from the membrane. Three prevalent models – shear-induced diffusion, inertial lift and the surface transport have been used to model the back transport of particles away from membranes. These models are valid only in the *laminar* flow regime.

3.3.5 Shear-Induced Diffusion

The shear-induced diffusion model was introduced by Zydney and Colton [84] as an alternative to the film model described earlier. In this shear-induced diffusion model, it was suggested that when subjected to a shear flow, the particles will randomly "bump into" and tumble over each other, which results in a random displacement of the particles. As such, the Brownian diffusivity (D) in Equation 3-6 of the film model is replaced by the shear-induced diffusivity to yield the following equation for flux:

$$J = 0.078 \gamma_o \left(\frac{a^4}{L}\right)^{1/3} \ln\left(\frac{\phi_w}{\phi_b}\right)$$
 [3-8]

This model suggests that flux is proportional to shear rate γ_0 by the power of 1 (as opposed to 1/3 in the film model), and proportional to particle size *a* by a power of 4/3. Increases in shear rate will have more of an effect on particle back-transport (and hence permeate flux) than is predicted by the film model. Increasing the particle size will also result in higher back-transport in the shear-induced model, while the opposite trend is predicted by the film model. Molecular diffusion becomes important for low shear rate and small particles, while shear-induced diffusion becomes more important for higher shear rate and larger particles [3].

3.3.6 Inertial Lift

Green and Belfort [83] suggested the inertial lift model in response to the "flux paradox" previously described. In the inertial lift model, a particle subjected to a laminar velocity distribution near the membrane surface undergoes rotation, and results in a lateral movement away from the membrane due to the differential pressure induced by the velocity gradient. The inclusion of the lateral migration of the particle away from the membrane due to inertial lift, yields the following equation for permeate flux [3]:

$$J = 0.036\rho \ a^3 \frac{\gamma_o}{\eta}$$
 [3-9]

In this inertial life model, permeate flux is proportional to shear rate by the power of 2, to particle size by a power of 3. This suggests that inertial lift becomes dominant at higher shear rate and larger particle sizes, compared to the shear-induced diffusion model.

3.3.7 Surface Transport Models

Surface transport models suggest that the mechanism of particle transport is not the backtransport mechanisms described by the inertial lift and shear induced models, but rather, particles at membrane surfaces roll along the surface due to tangential flow along the membrane surface. An approach to predict whether the particle will roll along the membrane or adhere to the surface is to consider the torque and forces acting on a single particle at the membrane surface. Forces acting on the particles are tangential drag force due to shear flow, and the normal drag force (relative to membrane surface) due to permeate flow. With gravitational force and adhesive force considered negligible, a torque balance is performed at the point of contact between the particle and the membrane surface. The force and torque balance then yield the following equation for the predicted permeate flux [3]:

$$J = 2.4a \gamma_o \left(a^2 R_c\right)^{2/5} \cot\theta \qquad [3-10]$$

All of the transport models described assume the following form, which relates flux to shear rate γ , particle size (a), concentration (ϕ), filter length (L) and suspension viscosity (η):

$$\left\langle J\right\rangle = c\gamma_o^{\ n}a^m\phi_b^{\ p}L^q\eta^r \qquad [3-11]$$

The constants c, n, m, p, q, and r vary for different models. The different models are suited for describing the back-transport phenomenon, depending on particle size and shear rate. Table 3-4 provides a summary of the values of the constants for the different particle transport models.

Table 3-4. Parametric dependence of the long-term flux for various transport mechanisms [4]

	Brownian diffusion	Shear-induced diffusion	Inertial lift	Surface transport
Shear rate (γ_o)	Increase	Increase	Increase	Increase
	n=0.33	n=1	n=2	n=1
Particle size (a)	Decrease	Increase	Increase	Increase
	M=-0.67	M=1.33	M=3	M=1
Feed concentration (ϕ_b)	Decrease	Decrease	No effect	No effect
	P=-0.33	P=-0.33	P=0	P=0
Suspension viscosity (η)	Decrease	Decrease	Decrease	No effect
	R=-1	R=-0.33	R=-1	R=0

The models described above are only valid for laminar flow regimes. Additionally, effects of particle electrostatic forces, compressibility, shape and size distributions were not considered in the models [49]. In a gas-sparged submerged hollow fiber membrane module, the flow regime is expected to be turbulent as a result of the complex hydrodynamic conditions induced by gas sparging. As such, these models developed are not likely applicable for studying fouling inside the submerged hollow fiber membrane modules. Moreover, as discussed in Section 3.2, several factors such as electrostatic interaction between particles, membrane properties, permeate flux or transmembrane

pressure, and feed characteristics also have an impact on fouling. This highlights the complexity in obtaining a representative fouling model for the submerged hollow fiber membrane module.

3.4 The Use of Air Sparging for Fouling Control

As previously discussed in Section 3.3.4, fouling control and reduction can be largely achieved through the reduction of solids on membrane surfaces. Several strategies have been developed to reduce solids concentration near the membrane surface, all of which involve controlling the hydrodynamics near membrane surfaces to increase the rate of particle back-transport away from membranes [3]. Some early developments in generating favorable hydrodynamic conditions on membrane surfaces for particle deposition control included placing protuberances on membrane surfaces to generate turbulent flow, using pulsating or oscillating flow in tubular membranes or using Dean or Taylor rotating filters to generate instabilities caused by Taylor or Dean vortices [3]. None of these strategies are commonly used for wastewater or water applications since they are associated with high operating and capital costs and some have been found to be inefficient in fouling reduction. A more recent development in inducing favorable hydrodynamic conditions is the use of air sparging or bubbling. A review of the existing literature on the use of air sparging or bubbling to enhance membrane processes is provided in the sections that follow.

The use of air sparging for fouling control in membrane processes has been gaining popularity over the past decade, particularly in the areas of drinking water production and biological wastewater treatment (i.e. membrane bioreactor) and macromolecular separation [85]. Air sparging has been widely acknowledged to reduce the extent of fouling by 30 to 300%, depending on the applications, operating conditions, membrane configuration (tubular, hollow fiber or flat sheet membranes) and characteristics of the liquid being filtered [56, 71, 86-90]. In this review, membrane configurations are classified into two categories: confined and unconfined modules. Confined modules (e.g.

tubular cross-flow membranes) are those for which the liquid to be filtered and sparged air bubbles are confined within the membrane. For these modules, the liquid typically permeates through the membrane in an inside-out flow configuration. Unconfined modules (e.g. submerged hollow fibers or flat sheet membranes) are those for which the membranes are submerged in the liquid to be filtered and then the membrane surfaces are scoured by sparged air bubbles. For these modules, the liquid typically permeates through the membrane in an outside-in flow configuration.

The mechanisms by which fouling control is achieved during air sparging is related to hydrodynamic conditions near the membrane surface [10, 47, 91]. Hydrodynamics, particularly shear stresses, have been recognized as an important parameter in controlling particle back-transport from membrane surfaces, increasing the rate of mass transfer and enhancing permeate flux [3, 10, 92-96]. In the discussions below, shear stresses induced by rising air-sparged bubbles are first reviewed for confined systems, so that the mechanisms by which foulant control is achieved in confined systems can be compared to those for unconfined systems.

For air-liquid two-phase flow inside a confined system, such as a tubular membrane or a hollow fiber, different flow patterns may exist. The most common upward two-phase flow patterns in vertical ducts are bubble flow, slug flow, churn flow and annular-mist flow, as illustrated in Figure 3-2. Bubble flow is characterized by continuous liquid flow with dispersed bubbles throughout a vertical duct. Slug flow occurs when smaller bubbles coalesce to a size of at least 60% of the duct diameter, flowing intermittently with liquid slugs [10]. Slug flows in confined tubular ducts generally exhibit the characteristics of a 'Taylor bubble', which are long bubbles with a rounded nose. Taylor bubbles rise in the center of a confined tubular duct and the liquid near the wall of the duct reverses its flow relative to the direction of the rising bubble (falling film region). Churn flow occurs when large bubbles form irregular shapes and the liquid oscillates around the large bubble, inducing mixing between the two phases. Annular flow occurs when the air phase is the dominant fraction in the duct and liquid flow is confined to the

liquid film on the duct's surface. As noted in the next section (Section 3.4.1), it is not clear whether all of the patterns seen in Figure 3-2 exists inside unconfined systems.



Figure 3-2. The different air-liquid flow patterns in a tubular vertical duct [87]

The different flow patterns described above have been reported to affect fouling control in confined membrane systems to different degrees, with slug flow consistently observed to minimize fouling on membrane surfaces to a greater extent than the other flow patterns [10, 87, 91, 97, 98]. Rising air slugs can be characterized in terms of four regions: the pre- (and post-) slug zones, the nose zone, the falling film zone, and the slug tail end zone. Ghosh and Cui [99] calculated the mass transfer coefficients induced on the surface of a vertical tubular duct by these four different regions using known empirical correlations, and found that the calculated mass transfer coefficients in the falling film and slug tail zones of a rising air slug were two orders of magnitude greater than those in the pre- and post-slug zones . Taha and Cui [97] used computational fluid dynamics

(CFD) modeling to study the distribution of wall shear stress induced by a rising air slug in a vertical tubular membrane. Similarly, Laborie and Cabassud [100] investigated the wall shear stress induced by an air slug flowing inside a rigid capillary tube using an electrochemical shear probe. Although the wall shear stress measured by Laborie and Cabassud [100] was non-directional, both studies reported shear stress patterns similar to that presented in Figure 3-3, which illustrates a CFD generated profile of the shear stresses acting on the surface of a tubular duct wall during the passage of one air slug. In point 1 (Figure 3-3), the wall shear stress is constant and its sign negative, since only liquid is rising in the vertical direction. When an air slug is introduced, the shear stress changes sign (point 2 in Figure 3-3) indicating flow reversal at the wall. The magnitude of the positive shear stress continues to increase in the falling film zone of the air slug until a plateau is reached when complete flow reversal is achieved (point 3 in Figure 3-3). At the tail end zone of the slug (point 4 in Figure 3-3) the shear stresses begin to fluctuate due to the unsteady nature of the wake that is present in this zone. The sudden abrupt burst of shear stress from a positive to a negative value indicates a change in direction of the liquid flow. In the post-slug zone (point 5 in Figure 3-3), the shear stresses once again becomes negative. The magnitude of the shear as a result of liquid flow is smaller compared to the magnitude of shear stress as a result of an air slug. A similar shear profile can be observed every time an air slug is introduced into the vertical duct.

Figure 3-4 shows the experimentally measured shear profile during the passage of a train of air slugs rising in a vertical tube. The sign convention of in Figure 3-4 is different that that in Figure 3-3. Similar to that seen in Figure 3-3, the passage of each air slug results in a change in the sign of the shear stress. The magnitude of the shear stresses measured during the passage of the liquid slug is also lower than the magnitude of the shear stress during the passage of the air slug.



Figure 3-3. CFD simulation of shear profile of a gas slug rising in a vertical tube Pipe diameter = 12.7 mm; Liquid flow rate = 1.0 L/min; Gas slug volume = 8.3 mL [97]



Figure 3-4. . Experimentally measured shear profile of a train of gas slugs rising in a vertical tube

Pipe diameter = 32 mm, gas superficial velocity = 0.3 m/s, liquid superficial velocity = 0.01 m/s [101]

3.4.1 Air Sparged Bubble Dynamics in Unconfined Systems

The preceeding discussion focuses on the behavior of slug flow in confined systems such as tubular membranes. Although the surface shear stresses induced by a rising airsparged bubble in a confined vertical duct have been extensively characterized both numerically and experimentally, very limited research has focused on characterizing the hydrodynamics of air-sparged bubbles in unconfined systems such as submerged flat sheet or hollow fiber membranes. This is in part due to the extremely complex nature of geometry and flow path of air-sparged bubbles in unconfined systems. Depending on the air sparging system, bubbles of different sizes and shapes can be introduced into an unconfined system. This relationship can be described by three dimensionless numbers the Reynolds number (Re), the Eotvos number (Eo) and the Morton number (Mo). The Re number represents the relationship between inertial forces and viscous forces, described in the following equation:

$$Re = \frac{\rho v D_b}{\mu}$$
 [3-12]

The Eo number represents the relationship between gravitational force and surface tension force, described in the following equation:

$$Eo = \frac{(\rho_L - \rho_g)gL^2}{\sigma}$$
 [3-13]

The Mo number represents the relationship between gravitational force and capillary force, described in the following equation:

$$Mo = \frac{g\mu^4 \left(\rho_L - \rho_g\right)}{\rho_L^2 \sigma^3}$$
 [3-14]

Figure 3-5 represents the correlation of the three dimensionless numbers described above to the shape regimes for bubble rising in an unconfined medium. Different bubble sizes and shapes also result in different terminal rise velocities (Table 3-5) and flow paths, and as a result, induce different shear stresses on the surrounding membrane surfaces.



Eo

Figure 3-5. Shape regimes for bubble rising in an unconfined media [102]

Table 3-5. Bubble types and rise velocities in stationary liquid [9]

Bubble Type	Bubble Size	Typical Terminal Rise Velocity
Spherical	< 1 mm	~ 0.1 m/s for 0.8 mm bubble
Ellipsoidal	1.5 – 15 mm	0.24 m/s for 4-15 mm bubble
Spherical Cap	> 15 mm	0.3 m/s for 20 mm bubble

Nagaoka et al. [103] measured the shear stresses acting on a wall adjacent to an airsparged flat sheet membrane using a two-direction force sensor. Two types of liquid matrices were used in their experiments: low viscosity liquid (1 mPa s) and high viscosity liquid (15 mPa s). Unlike the shear stress profile observed for confined systems (Figure 3-3), the time series data revealed that the shear stress profiles were characterized by short peaks in the magnitude of the shear stress. In addition, the profiles of these short peaks in shear stress were different for each shear event. Compared to the experiments in the high viscosity liquid matrix, the magnitude of the shear stress was found to be lower in the low viscosity matrix. For the low viscosity liquid matrix, the frequency of fluctuation of shear stresses (i.e. fluctuation in the direction of liquid flow near the surface of the wall) was higher than in the case for the high viscosity liquid matrix. As expected, these results suggest that fluid viscosity plays an important role in the hydrodynamic conditions experienced by the membrane surface during air sparging. Ducom et al. [93] measured the shear stress acting on flat sheet membranes during air sparging, using a relatively small non-directional electrochemical shear probe. Periodic shear events were observed over time, and these high shear stresses were attributed to air bubbles rising in close proximity to the shear probes on the membrane surface. Using CFD modeling, Ndinisa et al. [104] studied the shear stress distribution on an air-sparged flat sheet membrane submerged in water. Shear events were observed when air-sparged bubbles were present near the membrane surface. Also, at higher air sparging flow rates, their model showed that air bubbles migrated away from the membrane surface, inducing liquid up-flow between the membrane sheets and a liquid down-flow at the surface of the membranes. Higher shear stresses were observed when baffles were used, since air bubbles became more confined near membrane surfaces. The shear stresses induced by the interaction between sparged bubbles and the membrane surface is therefore dependent on the configuration of the membrane and the liquid matrix used.

For submerged hollow fiber membranes, there is limited information available on the bubble induced shear stresses imparted onto membrane surfaces. As a result, some researchers have assumed that since the packing density of fibers in a submerged

membrane system is relatively high, the shear stresses imparted onto membrane surfaces by air-sparged bubbles rising between fibers is similar to that induced by an air slug in a confined system [10, 105, 106]. However, the validity of this assumption is questionable, given the complex and ever-changing nature of the hydrodynamic conditions in submerged hollow fiber membrane systems. In submerged hollow fiber membrane systems, the fibers are normally held loosely and typically sway extensively. This irregular fiber configuration continuously changes the flow path of rising air-sparged bubbles. Bérubé et al. [107] measured the shear stresses on the surface of hollow fibers in a submerged membrane system using a non-directional electrochemical probe. Typical surface shear measurements obtained from this study are illustrated in Figure 3-6. As presented, the frequency, duration and amplitude of the shear events were highly variable, and significantly different from the shear profiles of air slugs rising in a confined tubular system. The electrochemical shear probe developed by Bérubé et al. [107] was only capable of measuring the magnitude of the shear stresses, not the direction of liquid flow, and therefore could not be used to comprehensively characterize the effect of rising air bubbles in a submerged membrane system. Nagaoka et al. [108] used a two-direction load sensor to record the shear stress profile acting over the entire length of the hollow fiber during gas sparging. Their reported profiles were similar to those observed by Bérubé et al. [107], i.e. highly variable frequency, duration and amplitude of shear events. However, their reported magnitudes of shear stresses were up to ten times higher than those reported by Bérubé et al. This may be due to the fact that the two-direction load sensor used by Nagaoka et al. measured the shear stress acting on the entire length of the fiber, while the electrochemical shear probe used by Bérubé et al. measured locally the shear stress acting on a small section of the fiber. Moreover, the sensitivity of the shear instrument presented by Nagoaka et al. was low, and does not offer insight into shear stress induced during the passage of a single bubble. Yeo et al. [109] used particle image velocimetry (PIV) to measure liquid velocities in an air sparged hollow fiber module. They observed that the axial velocities inside a hollow fiber module were up to ten times lower than those outside of the module where sparged air bubbles were introduced. This observation is similar to those of Berube et al. [107] who suggested that "a tightly configured multi-fiber module could potentially shield certain areas of a fiber from both the bulk liquid flow and sparged gas bubbles" [107].

Based on the above discussions, the hydrodynamic characteristics, and therefore, the mechanisms by which air sparging reduces the extent of fouling, is expected to be substantially different for confined and unconfined membrane systems. Consequently, the assumption that shear stresses imparted onto membrane surfaces by air-sparged bubbles in submerged unconfined systems are similar to those in confined systems is likely not valid.

Despite the investigations by Nagaoka, Berube and Yeo, much remains unknown about the impact of membrane module configuration, and gas sparging practices on the hydrodynamic conditions inside submerged hollow fiber membrane module. Moreover, shear profiles inside submerged hollow fiber modules are characterized by highly variable, oscillatory shear condition (see Figure 3-6). Variable, transient shear condition have been found to enhance fouling control by enhancing particle transport away from the surface [110], or detaching the biofilm on the surface of the membrane [111]. However, it has been shown that under variable, oscillatory flow conditions, there exists a critical oscillation frequency, above which the flow condition becomes detrimental to the lift forces that transport particles away from the membrane as a result of secondary flow effects [110]. It remains unknown to date what the optimum frequency of oscillation is in terms of fouling control inside the gas-sparged submerged hollow fiber membrane module.



Figure 3-6. Profile of shear stresses acting on a submerged hollow fiber membrane under continuous aeration [107]

3.5 Mechanism of Fouling Control and/or Permeate Flux Enhancement by Air Sparging

Although the exact mechanisms by which air sparging enhances the permeate flux is not known, many possible mechanisms have been suggested in the literature. These suggested mechanisms are summarized in this section. It is noted that the studies which investigated the possible mechanisms were mostly based on confined systems in which the hydrodynamic conditions are well defined and understood. As discussed in Section 3.4, the hydrodynamic conditions inside the gas-sparged submerged hollow fiber membrane module are not well understood. As such, it is not clear to what extent these mechanisms occur inside the gas-sparged submerged hollow fiber membrane module.

Physical Disruption of the Concentration Polarization Layer

Concentration polarization, which can occur at the surface of a membrane due to the accumulation of retained solutes, limits the overall mass transfer at a membrane surface, and therefore increases the overall resistance of a membrane to permeate flow. Bellara et

al. [56] suggested that in a confined system, a rising air slug could scour the membrane surface, reducing the thickness of the mass transfer-limiting layer and therefore, the overall resistance of a membrane. Using CFD modeling, Smith et al. [112] and Taha and Cui [97] also reported that the flow reversal that occurs at the falling film region of a rising air slug in a confined system had the greatest beneficial impact on reducing the thickness of the mass transfer-limiting layer. However, the effective scouring that results from the falling film can only be expected if a rising air bubble is in close proximity to a membrane surface, as observed for air slugs in confined systems. For small air bubbles in confined systems, the rising air bubbles may not come in close contact with the membrane surface. Under these conditions, the impact of rising air bubbles on the thickness of the mass transfer-limiting layer is expected to be affected by the proximity of a bubble to the membrane surface. This was observed experimentally by Li et al. [113] when investigating the effect of air bubble sizes on the permeate flux in a confined system. For small air bubbles, the permeate flux that could be achieved was lower than that which could be achieved for larger air bubbles. A maximum achievable permeate flux was observed when the diameter of a rising air bubble was similar to that of the diameter of the tubular membrane (i.e. slug flow). Further increasing the size of the gas slug was found to have no effect on the size of the wake [114], and therefore did not have an impact on the permeate flux.

The degree of flux enhancement due to concentration polarization disruption also varies depending on the severity of flux decline. For example, Cui and Wright [91] and Bellara et al. [56] observed that flux enhancement was the greatest when concentration polarization was most severe, and that flux enhancement was greater when the dominant mechanism of flux decline was concentration polarization, as opposed to cake formation or internal fouling. Flux decline due to concentration polarization is more reversible than cake formation and internal fouling. Fouling control in systems where cake formation is the dominant mechanism is likely due to the modification and/or erosion of the cake structure, as discussed later.

In unconfined systems, it is not clear if the mechanism of membrane surface scouring through the falling film surrounding a rising bubble occurs, and if its contribution to fouling control is significant relative to other possible mechanisms. Effective scouring in unconfined systems is also only expected if rising air-sparged bubbles are in close enough proximity to the membrane surface to generate flow reversal. Lee et al. [115] and Ndinisa et al. [116] observed improvements in fouling control during filtration using flat sheet membranes by decreasing the width of the flow channel for two phase flow. However, it was not clear if the improvements were the result of scouring by the falling film surrounding the bubble (which reduces the thickness of the mass transfer-limiting layer [97]), or the result of enhanced mass transfer of particles away from the membrane surface due to increased shear stresses near the membrane surface.

Bubble-Induced Oscillatory Flow

In confined systems, a rising air slug generates secondary oscillating flows in the wake of the tail end zone. These secondary flows result in relatively high shear stresses of short duration. These high shear stresses likely prevent the accumulation of retained material on membrane surfaces [3, 47, 56, 87, 98], and/or reduce the thickness of the mass transfer-limiting layer [112, 116]. Yeo et al. [109] observed a strong correlation between the extent of fouling control and the standard deviation of the liquid velocity along a membrane surface. Typically, as the standard deviation in the liquid velocity increased, the extent of fouling control increased. Increases in shear stresses have also been correlated with fouling control in air-sparged submerged membrane (i.e. unconfined) systems [93, 117]. As previously discussed, the shear stresses in these systems are highly variable, indicating the presence of oscillating flows near membrane surfaces [103, 118]. Oscillatory and transient shear stress was also reported to lead to biofilm detachment on a surface. It is probable that these oscillating flows induced by rising air-sparged bubbles are the source of significant fouling control for unconfined membrane systems. However, as discussed in Section3.3, there could exist a critical frequency of oscillation, above which the oscillatory flow may be detrimental to fouling control.

Some researchers have attributed the higher permeate flux that can be maintained when using air sparging, to the bulk liquid flow induced by rising air-sparged bubbles [10]. However, for a given bulk cross flow velocity in an unconfined system, Bérubé and Lei [86] observed that the permeate flux that could be achieved was substantially greater for two-phase flow (i.e. with air sparging) than for single-phase flow (i.e. without air sparging) in the submerged hollow fiber module. In addition, Bérubé et al. [107] observed that for a given bulk cross flow velocity, the magnitude of both the average and peak surface shear stresses were substantially greater for two-phase flow. Similar observations were made by others when using flat sheet membrane [104] and tubular membranes [119]. Therefore, the overall contribution to fouling control from the bulk liquid flow induced by rising air-sparged bubbles, may not be significant for unconfined system, when compared to other mechanisms.

Modification of Cake Structure

The bubble-induced oscillatory flow discussed above, has been linked to modifications of the cake structure already formed on a membrane surface, such that resistance to flow is decreased and flux is enhanced. Mercier-Bonin et al. [120] suggested that the destabilization or erosion of the cake layer formed on the membrane surface by the imposed shear stress was the primary mechanism of fouling control. Laborie [121] and Cabassud [47] reported that a cake structure can be drastically modified by gas bubbles passing by the membrane surface during filtration. Increasing gas flow rate results in increased cake porosity and a decrease in cake specific resistance. However, there exists a critical gas flow rate above which cake porosity decreases and cake specific resistance increases.

Modification of the Biofilm Property

For membrane foulants that include a biofilm, an imposed shear stress near the membrane surface can have an effect on the biofilm community. Rochex et al. [122] investigated the effect of sher stress on a biofilm, and they found that the composition of the bacterial community in the biofilm, as well as the biofilm diversity are impacted by shear stresses. They also suggested that shear stress would "*slow down biofilm maturation and tend to*

maintain a young biofilm." This may lead to a reduced biofilm adhesion, thus making it easier for the biofilm to detach from a membrane surface. Additionally, Ochoa et al. [111] found that non-uniform, variable shear stress imposed near the biofilm is much more effective in detaching the biofilm than a constant shear flow imposed on the membrane surface. These observations suggest that non-uniform, variable shear stress plays an important role in the removal of biofilms on membrane surfaces.

Pressure Instabilities

Cui and Wright [89] suggested that the pressure instabilities caused by a rising air slug can have beneficial effects on the permeate flux in confined membrane systems. These pressure instabilities are caused by relatively high pressure zones that are present at the nose end and tail end of air slugs, respectively. Mercier et al. [123] suggested that the degree of fouling control depends on the time required for a cake layer to be formed on the membrane surface, and the frequency of the pressure instabilities. If the frequency of pressure instabilities (or bubble period) is higher than the inverse of the cake formation time, then prevention of cake formation on the membrane surface could be achieved.

3.6 Influence of Module Configuration and Operating Conditions

Shear stresses generated by air sparging are affected by the sparging flow rate, the sparging intensity, the geometry of the sparged bubbles and the membrane module configuration. This section provides a brief review of the impact of these parameters on hydrodynamics of submerged air-sparged membrane systems.

3.6.1 Sparging Intensity and Bubble Geometry

The mechanisms of mass transfer boundary layer thinning and the high shear stresses generated in systems with two-phase flows suggest that the benefits of air sparging are largely dependent on the number of shear events produced by rising bubbles. Therefore, increasing sparging intensity should increase the number of shear events experienced by the membrane [124]. Ghosh and Cui [99] observed that increasing air sparging intensity, which increased the number of shear events, improved fouling control in a confined membrane system. A similar trend was observed by Bellara et al. [56] and Cabassud et al. [47]. Shear stresses generated by a rising air bubble in a confined system also depend on the size of the bubble and the confinement area that the bubble is rising through. Although longer air-sparged bubbles have been suggested to provide better fouling control [113], the positive effect of the size of air slugs appears to be dependent on the type of membranes used. Smith et al. [112] suggested that the shear profile of slug flow inside a hollow fiber is different from that of the profile inside a tubular membrane, where many of the spikes in the wall shear stress were only observed at the nose and tail end of the bubble, not in the falling film region. In addition, for a given air sparging flow rate, increasing the bubble size decreases the frequency of shear events generated by air sparging, potentially decreasing fouling control [118].

In unconfined systems, the effects of shear induced by gas sparging was found to be beneficial, but only up to a critical point. Khan et al. [125] studied the effect of shear intensity on fouling rates in a bench-scale MBR. Different shear intensities were created through mechanical stirring in the reactor. Initially fouling rates decreased with increased shear intensity, until an optimum shear intensity was reached. Above the optimum shear intensity, fouling rates increased. Ueda et al. [71] investigated the effects of air sparging intensity in an unconfined membrane system. They observed that concentrating the diffusers used for air sparging in a submerged hollow fiber system to a smaller area, and thus increasing the aeration intensity, resulted in a reduction in membrane fouling. They also reported that although increasing the air sparging flow rate generally decreased the extent of fouling, a critical air sparging flow rate was observed above which further increasing the flow produced no further decrease in the extent of fouling. Similar observations have been reported by Bouhabila et al.[126], Bérubé and Lei [86] and Ghosh [127] for submerged hollow fiber modules, and Ndinisa et al. [116] for submerged flat sheet membranes. These observations suggest that a critical air sparging flow rate exists above which further increases in the sparging flow do not result in an increase in shear stress on submerged membrane systems. This is consistent with

the observations by Bérubé et al. [107], who reported that at a high air sparging flow rate, an incremental increase in the air sparging flow rate resulted in a lower increase in the surface shear than an incremental increase at low air sparging flow rates. Several hypotheses are available to explain this phenomenon. Firstly, at high air sparging flow rates, sparged air bubbles tend to coalesce. This was observed experimentally by Nguyen et al. [128], who, using a bi-optic probe to characterize bubble diameter and velocity inside a commercial gas sparged submerged hollow fiber module. The coalescing of the bubbles will result in decreased number of shear events experienced by membrane surfaces. Secondly, the effective membrane area available for filtration can also decrease if air bubbles occupied a significant portion of a membrane surface at high sparging rates [129]. Thirdly, particle deposition on membrane surfaces is dependent on back-transport mechanisms (i.e. shear stresses). At the critical air sparging rate, the shear stresses controlling particle back-transport are high enough to prevent any particle deposition on membrane surfaces. As such, a further increase in sparging rate (and therefore shear stress) has no additional effect on prevention of particle deposition and reversible fouling [116].

In submerged hollow fiber membranes, Yeo et al. [109] and Fane et al. [130] reported better fouling control when sparging with smaller air bubbles, compared to larger ones. This is likely related to the fact that for a given air sparging flow rate, sparging with smaller bubbles results in a higher number of shear events, compared to sparging with larger bubbles. However, sparging with larger bubbles can potentially generate shear events of greater magnitude [104, 131]. Therefore, the selection of the optimum sparged air bubble size potentially depends on the competing benefits of an increased numbers of shear events which are induced by smaller air bubbles and shear stresses of larger magnitude induced by larger air bubbles [131]. Sofia *et al.* [132] examined the effects of diffuser type (fine vs. coarse) on fouling control in flat sheet membranes. When compared to coarse bubbles (2 mm diffuser hole), sparging with fine bubbles (0.5 mm diffuser hole) resulted in better fouling control. However, Ndinisa et al. [116] observed the opposite trend for flat sheet membranes, for which better fouling control could be achieved when sparging with larger bubbles. This discrepancy can be related to the size

of the bubbles relative to the width of the flow path between the membranes. The width of the flow path in the system used by Ndinisa et al. was 7 mm. Therefore, bubbles greater than 7 mm in diameter were essentially confined and the resulting falling film potentially scoured the surface of the membranes [104]. The width of the flow path and the bubble size in the system used by Sophia et al. [132] is not known. However, based on their results, it is likely that the diameter of the larger bubbles was smaller than the width of the flow path. Prieske et al. [3, 133] also suggest that larger bubbles can be more efficient in fouling control compared to smaller bubbles, because larger bubbles can induce a higher recirculation velocity within the flat sheet membrane module. The optimal bubble size in a given membrane module is also dependent on module configuration such as channel gap width between membranes. Inside a submerged hollow fiber membrane module, further complication arises where different fiber packing densities and gas sparger design can result complex bubble – fiber bundle interaction. This interaction can lead to different hydrodynamic conditions that are generated at the membrane surface.

3.6.2 Membrane Configuration

The hydrodynamic conditions experienced by air-sparged submerged membrane fibers are dependent on the location of the fiber within the membrane module, and the fiber packing density. Fibers that are located in the outer sections of a module of hollow fibers are expected to experience the benefits of the hydrodynamic conditions generated by a rising sparged air bubble to a greater extent than fibers that are located within the module, especially if the packing density is high. Kiat *et al.* [134] reported higher inter-fiber fouling for hollow fiber bundles when the fiber packing density was greater than 20 fibers/cm². Chang and Fane [52] suggested that for high fiber packing density modules, sparged air bubbles rising between fibers were smaller and therefore had slower rise velocities than bubbles in a module with a low packing density. Similarly, Yeo and Fane [135] studied the effects of fiber packing density on the performance of the individual fibers in a bundle of submerged hollow fiber membranes, and reported greater fouling control when the packing density was reduced to less than a critical value. At higher

packing densities, the performance of the fibers (in terms of permeability) located in the center of the bundle was very low, due to the variation of hydrodynamic conditions experienced by different fibers in the bundle. These results indicate that densely packed fibers likely dampen the effects of increased crossflow caused by rising bubbles, or prevent the rising bubbles from reaching the inner sections of the fiber bundle.

Yeo and Fane [135] found that fibers within the fiber bundle fouled more quickly, compared to a single fiber under the same experimental conditions. The higher fouling rate was attributed to "stagnant" flow areas within the bundle. This "stagnant" area or shielding effect can potentially lead to inter-fiber fouling, where cake growth on individual fibers began to merge with the cake layers on other fibers, resulting in several fibers sticking to each other through the cake layer [134, 135]. Nagoaka [108] and Yeo et al. [109] found that the hydrodynamics experienced by fibers inside a bundle is different than that of fibers outside of the bundle. Yeo et al. used particle image velocimetry (PIV) to measure liquid velocities in an air sparged hollow fiber bundle. They observed that the axial velocities outside a hollow fiber bundle were up to ten times higher than those inside of the bundle where sparged air bubbles were introduced. Yang et al. [136] also suggested that individual fibers within a bundle may experience "permeate competition" from surround fibers. This permeate competition contributes to an overall hydrodynamic resistance during filtration.

The distribution of a concentration polarization layer along the fiber length may be dependent on the fiber length, as demonstrated by Carroll and Booker [137]. They suggested that pressure drop in the fiber lumen is the culprit for this axial distribution of fouling. Similarly, axial distribution of fouling along the fiber length is also reported by Yoon et al. [138], who showed that transmembrane pressure is highest at the suction end of the fiber. This resulted in faster fouling at the suction end of the fiber. However, it is possible that the increased fouling rate at the suction end of the fiber can be somewhat mitigated by the increased number of shear events at the top end of the vertically aligned fiber. Nguyen et al. [128] found that there is a higher number of bubbles at the top of the submerged fiber module than at the bottom of the fiber module. Increase number of bubbles may result in increased number of shear events at the membrane surface.

Additionally, in a gas sparged module, bubbles at the top of the module can be larger than the bubbles at the bottom of the module, due to the coalescense of bubbles, as well as lower pressure at the top end of the module compared to the bottom of the module. Larger bubbles may generate higher shear stresses, which can be beneficial for enhancing particle back-transport from membrane surfaces. Using an electrochemical probe, Berube et al. [107] confirmed that highest shear stresses were indeed measured at the top end of the vertically aligned hollow fiber during gas sparging.

Chang and Fane [139] investigated the effect of fiber diameter on a submerged hollow fiber module. They observed that air sparging had a more beneficial impact on fouling control for fibers with a smaller diameter than those with a larger diameter. They hypothesized that fibers with a smaller diameter were capable of swaying to a greater extent than those with a larger diameter, and that this enhanced fouling control. Similarly, Wicaksana et al. [140] observed that fiber movement was greater for fibers with smaller diameters. The benefits of loosely held hollow fibers in submerged systems have been known for some time, and a certain degree of looseness is known to yield better fouling control, compared to tightly held fibers. In fact, many industrial membrane manufacturers (e.g. GE-Zenon, Siemens, Pall) operate their submerged membrane systems based on a loosely held configuration [10]. Chang and Fane [52] reported that fouling was more extensive for tightly held fibers compared to loosely held ones. Similarly, Bérubé and Lei [86] and Martinelli [141] observed that significantly better fouling control could be achieved for the loosely held fibers, compared to the tightly held fibers. Wicaksana et al. [142] reported that the critical flux for loosely held fibers was higher than that for tightly held fibers when filtering a yeast suspension. Both Chang and Fane [52] and Wicaksana et al. [142] attributed the improvement in fouling control to the ability of loosely held fibers to sway. Wicaksana et al. [140] also examined the effect of fiber movement on the extent of fouling by mechanically moving the fibers to simulate fiber movement. Compared to conditions in which the fibers did not move, mechanically moving the fibers resulted in a reduced fouling rate. When combining mechanical movement and air sparging, the rate of fouling was even less than just mechanical movement, suggesting that both fiber movement and shear events contribute to fouling
control [140]. Bérubé and Lei [86] suggested that in addition to the lateral movement of the fibers, loosely held fibers could also physically contact each other, potentially scouring the membrane surface and removing accumulated foulant. Also, Bérubé et al. [107] measured the shear stress acting on air-sparged submerged hollow fiber membranes and observed a higher frequency of shear events for loosely held fibers, when compared to tightly held fibers. The higher frequency can potentially lead to increased fouling control during filtration. However, it is possible than excessive physical contact may lead to membrane attrition over time. Therefore, there may be an optimum degree of fiber looseness, when weighing the benefits of reduced fouling versus increased cost of membrane replacement over time.

3.7 Linking Shear to Fouling Control

The benefits of air-sparging induced shear stresses on fouling control are evident, as discussed above. However, the extent of this benefit is also dependent on the membrane geometry and operating conditions. Unfortunately, to date, there have been no methods presented to systematically quantify the benefit of shear stresses on fouling control for different types of membranes under different operating conditions. Therefore, it is difficult to design and build a full scale submerged unconfined membrane module without resorting to the capital and time intensive process of pilot testing to obtain the required design parameters before a full-scale membrane module can be designed.

Several researchers have attempted to quantify the relationship between shear stress and fouling control. All studies conducted were performed using a lab-scale membrane modules. Leberre et al. [94] suggested the use of an effective shear stress in characterizing the "competition between convection and erosion at the membrane/solution interface." The effective wall shear stress was defined as :

$$\tau = \tau_w - \tau_w^* \qquad [3-15]$$

The critical erosion shear stress was determined experimentally. When τ_w is less than τ_w^* there is no transport of particles away from the cake, i.e. no cake erosion. The definition of effective shear stress was applied for constant shear conditions, and no consideration was given to oscillatory flows observed in two-phase gas sparging conditions.

For transient shear stresses in mechanically vibrating membrane modules, several researchers proposed the following relationship between permeate flux and the time averaged *mean* wall shear rate [92, 143]:

$$J = a\gamma^{-n}$$
 [3-16]

Jaffrin [144] proposed the same relationship, but instead of using a time-averaged mean shear rate, a *maximum* shear rate was proposed. The authors found in their experiments that flux was governed by the maximum shear.

To take into account the oscillatory flows induced by the passage of liquid and gas slugs in a confined membrane, Cabassud et al. [47] proposed three hydrodynamic parameters to link shear and flux enhancements. These relationships were based on their experimental investigation of ultrafiltration inside a gas sparged hollow fiber membrane:

i. a mean shear stress that takes into account the rate of occurrence of the gas slug

$$\overline{\tau} = \beta \tau_{gas} + (1 - \beta) \tau_{liq} \qquad [3-17]$$

ii. a total shear stress, and

$$\tau_{tot} = \tau_{liq} + \left| \tau_{gas} \right|$$
 [3-18]

iii. a pulsatile Reynolds number that accounts for oscillating flow in a cylindrical tube

$$\operatorname{Re}_{puls} = \frac{\rho_L D2\pi f_p A}{\mu_L}$$
 [3-19]

where A is defined based on the lengths of the liquid and gas slugs as:

$$A = \frac{L_L + L_g}{2}$$
 [3-20]

When trying to establish the relationship between these three hydrodynamic parameters and flux enhancement, no good correlation was found between flux enhancement and the mean shear stress τ_{mean} . However, a good correlation was found between flux enhancement and total shear τ_{tot} and pulsatile Reynolds number Re_{puls} . The authors noted that there was no physical significance in correlating flux enhancement to total shear (i.e. in a tubular membrane subjected to both liquid and gas slug, a particle is not only subjected to the positive shear stress, but also the negative shear stress induced by the gas slug.) The alternating positive/negative shear stresses may be responsible for inducing oscillatory movement of the particle. The authors did suggest that τ_{tot} and Re_{puls} can be correlated to a mixture Reynolds number. This suggests that flux enhancement may be "linked to mixing or turbulence near the membrane surface, more than mean shear stress" [47].

Based on their experimental measurements of wall shear stress and flux enhancements during air-sparged filtration using flat sheet membranes, Ducom et al. [93] also proposed the following hydrodynamic parameters to link shear stress and flux enhancements:

i. the ratio of two-phase time-averaged shear stress with sparging to wall shear stress with without air sparging,

$$\frac{\overline{\tau_{\text{two-phase}}}}{\overline{\tau_{\text{single-phase}}}}$$
[3-21]

ii. the ratio of the amplitude of the wall shear stress with sparging to wall shear stress without air sparging,

$$\frac{\left(\tau_{\max} - \tau_{\min}\right)_{\text{two-phase}}}{\tau_{\text{single-phase}}} \qquad [3-22]$$

iii. the frequency of bubbles.

For their experiments, Ducom et al. [93] found a good correlation between parameters defined in equations 3-21 and 3-22 and flux enhancement. This suggested the importance of transient shear stresses in limiting fouling. However, no correlation could be established between frequency f and flux enhancement. Yeo et al [184] demonstrated a good correlation between standard deviation and the rate of transmembrane pressure increase during filtration.

3.8 Summary

Membrane fouling is a costly problem in filtration processes. Many factors affect reversible membrane fouling: feed water characteristics (e.g. particle size and shape, particle electrostatic interaction), membrane property (e.g. pore size and hydrophobicity), and operating conditions (e.g. induced hydrodynamic conditions, operating pressure and flux). There are numerous studies in the literature that investigate the relationship between these factors and fouling. However, several contradictory observations were reported. One main reason for these contradictions is that each study was conducted under different operating conditions as well as membrane geometry. This highlights the importance of understanding the underlying mechanisms of fouling. Several models have been developed in an attempt to understand fouling behavior on membrane surfaces. The models can be classified into three categories: (1) resistance-in-series models, (2) flux decline models, (3) film model and (4) particle transport models. Though useful for a controlled study, these models are limited to simple and ideal conditions, and cannot used to model real-life membrane operations.

A popular strategies used to limit membrane fouling is to control of hydrodynamics through the use of air sparging. Several mechanisms by which air sparging limit fouling have been hypothesized. However, these mechanism can not be confirmed, especially in a submerged hollow fiber membrane system, since the hydrodynamic conditions surrounding the hollow fiber as a result of bubbling flow and fiber movement are poorly understood. Some researchers have assumed that the shear stresses imparted onto membrane surfaces by air-sparged bubbles rising between fibers are similar to that induced by an air slug in a confined system. However, the paths of bubbles inside submerged hollow fiber membrane module are very different than those inside a tubular membranes, especially since the fibers can sway extensively during air sparging. Therefore, the hydrodynamic characteristics are expected to be substantially different for confined and unconfined membrane systems.

In submerged hollow fiber modules it is difficult to control the hydrodynamic environment between the fibers during gas sparging. The sparging flow rate, the sparging intensity, the geometry of the sparged bubble and the membrane module configuration affect the hydrodynamic conditions surrounding each fiber membrane. Contradictory observations have been reported regarding the effect of bubble size on membrane fouling. These contradictions are likely due to the different operating conditions and membrane geometry used in each study, all of which have an impact on the shear stresses near a membrane surface. This highlights the importance of systematically quantifying the benefits of shear stresses on fouling control for different types of membranes under different operating conditions.

Very limited studies are available that experimentally investigate the hydrodynamic conditions, and more specifically the shear stresses inside a submerged hollow fiber membrane system. Only three studies – one using PIV for a hollow fiber bundle (Yeo and Fane), one using two-direction load sensor (Nagaoka et al.) and one using the electrochemical shear probe for hollow fiber bundle (Berube et al.) have been found. The PIV method employed by Yeo and Fane has a line-of-sight limitation, and cannot report

shear stresses inside a hollow fiber bundle. The two-direction load sensor employed by Nagaoka has a relatively low resolution, and is only capable measuring shear stress acting over the entire length of the fiber. The electrochemical shear probe employed by Bérubé et al. does not have a line-of sigh-limitation, is non-intrusive and has a relatively high resolution. However, the probe developed is only capable of measuring the magnitude, not the direction, of the shear stress. Therefore the occurrence of flow reversal near a membrane surface during the passage of an air-bubble can not be captured.

The experimental studies described in the subsequent chapters in this thesis are dedicated to investigate (1) the nature of the hydrodynamic conditions in submerged hollow fiber membranes during various gas sparging conditions, through the measurement of shear stresses acting on membrane surfaces, and (2) the fundamental relationship between shear and fouling rate. An electrochemical probe capable of measuring both the magnitude and the direction of shear stresses was developed for the investigations. Knowledge gained from this is paramount in understanding the mechanism of fouling inside submerged membrane systems, as well as optimizing the system from an energy perspective.

3.9 Identified Knowledge Gap and Thesis Direction

The hydrodynamic conditions, and more specifically, the shear stresses imparted on the hollow fiber as a result of bubbling flow and fiber movement are poorly understood. This knowledge gap presents a real impediment to understanding fouling behavior on membrane surfaces, and to optimizing the performance of submerged hollow fibers modules in terms of membrane geometry and energy costs associated with air sparging. As a result, an expensive and time-consuming process of pilot-testing is always required to obtain the design parameters before a full-scale system can be built.

The overall objective of this proposed research is to address the knowledge gap discussed above from the point of view of hydrodynamics, thereby providing a portion of the information necessary for developing a complete model of the submerged hollow fiber filtration process for water and wastewater applications. In order to achieve the objective, the following questions will be addressed:

- 1. What is the typical shear profile inside an unconfined system under gas sparging, and is the hydrodynamic condition near membrane surfaces of unconfined system similar to that of confined system? Shear stresses acting on the membrane surface have been known to have a direct impact on flux enhancement during filtration processes. Although shear stresses in a confined system such as tubular membrane, are well documented, the shear stresses acting on membrane surfaces of outside-in hollow fibers modules (i.e unconfined) are not well understood. Many researchers assume that the shear stresses acting on unconfined membranes are similar to those in a confined system, and the question herein attempts to address this assumption.
- 2. In an unconfined system what mechanisms contribute to the peaks in shear stresses observed (as seen in Figure 3-6). For example, are the peaks the result of the physical contact between fibers, the result of the falling film and turbulent wakes generated by a rising bubble or due to the lateral flow of the fluid induced by swaying fibers?
- 3. How do gas sparging practices (i.e. diffuser design and sparging intensity) impact the distribution of shear stresses inside a bundle?
- 4. How do different types of shear profiles examined in unconfined systems affect the degree of fouling control?

In answering the questions above, two types of experiments were performed in this thesis. The first type of experiment was dedicated to examining the hydrodynamic shear profiles inside a submerged hollow fiber membrane module when subjected to gas sparging. The placement of the gas sparger relative to the membrane fiber were based on two types of commercially available submerged hollow fiber membrane module – one

with sparging within a bundle (i.e. Puron membranes), the other with sparging outside the bundle (i.e. ZW 500 membranes). Chapter 5 and 6 investigated the typical shear profile when gas sparging occurred within a bundle, while Chapter 7 investigated typical shear profile when sparging occurred outside a bundle. Gas sparger sizes of 1 mm and 3 mm were used in the experiments. These sizes were similar to the gas sparger sizes adopted in commercial membrane systems (3 mm to 5mm), as well as those used by other researchers in their studies. In Chapters 5 and 6, gas flow rates ranging from 2 to 35 mL/min were investigated. These gas flow intensities were similar to those used by other researchers in their studies. In Chapter 7, the gas flow rates ranged from 10 to 195 mL/min. The higher gas flow rates were chosen to investigate whether or not a critical gas flow rate exists, above which no further increase in shear stresses were observed by some researchers.

The second type of experiment was dedicated to examining the relationship between shear profile and fouling. In Chapter 8, filtration experiments were conducted under controlled hydrodynamic conditions similar to those observed in Chapters 5, 6 and 7. The water matrix used was bentonite and water. Only reversible fouling was examined since the smallest bentonite particle was larger than the pore size of the membrane.

In order to perform the experiments described above, a tool which is capable of measuring shear profiles inside the submerged hollow fiber membrane module must be developed. Chapter 4 describes the development of the electrochemical shear probes, which were used to quantify shear profiles in the experiments in Chapters 5 to 8. If the reader is not interested in the detailed development of the shear probe, then he/she may proceed directly to Chapter 5, and bypass Chapter 4 altogether.

4. Wall Shear Stress Measurement Using the Electrochemical Method

4.1 Introduction

The characterization of hydrodynamic conditions near a membrane surface is important for understanding the mechanisms of fouling during membrane filtration. Although the electrochemical shear probes have been used extensively to quantify hydrodynamic conditions in confined systems such as tubular membranes, their ability to do so in submerged hollow fiber membrane modules during gas sparging has not been comprehensively investigated. To date, there is limited knowledge available with regards to the hydrodynamic conditions that exist inside gas sparged submerged hollow fiber membrane modules.

The aim of this chapter is to develop an electrochemical shear probe capable of measuring both the magnitude and the direction of shear stress imparted on a hollow fiber membrane surface during gas sparging. Two shear probe systems were developed – one on the inner wall of a rigid vertical tubular pipe, and the other on a non-rigid outer wall of a small diameter tube. The small tube was similar in geometry and flexibility to that of a hollow fiber membrane. The experiments were conducted for both single-phase and two phase flow conditions. Validation of the shear probes was achieved by comparing the experimentally measured shear stress values to theoretically calculated shear stress values (single-phase conditions), and to known shear profiles (two-phase flow conditions) in the literature. These known profiles were obtained using CFD modeling as well as experimental approaches (for two phase flow conditions) for confined systems.

This chapter describes the shear probes developed, and the validation procedure used to ensure that the probe can provide accurate measurements of both the magnitude and direction of shear stress imparted on a membrane surface. The probe developed in this chapter was then used in subsequent experiments (described in Chapters 5 to 8) to characterize hydrodynamic conditions inside bench-scale gas sparged submerged hollow fiber modules.

4.2 Background

An important parameter in studying fouling control is the shear stress acting on the membrane surface. The measurement of the wall shear stress using electrochemical methods was first performed by Reiss and Hanrraty [145], and has been used extensively by other researchers to study mass transfer and local hydrodynamic properties in fluidized reactors, two phase pipe flows etc. (e.g. [146-149]). The principle of the electrochemical method for the measurement of wall shear stress, is that fluid flow depends on the rate of diffusive mass transfer between a cathode and an anode in an electrochemical cell. In this section, the principles of the electrochemical shear method are first discussed. The use of the electrochemical shear method by others in investigating shear stresses in pipe flow, and in membrane systems during gas sparging is then provided.

4.2.1 Principle of the Electrochemical Shear Method

When a potential difference is applied between two electrodes immersed in a stagnant solution of reacting species, a relationship between the applied potential and the measured current density in the electrochemical cell similar to that seen in Figure 4-1, is observed. In zone I, the reaction rate at the electrode is less than the rate of arrival of the reacting species to the electrode. Increasing applied potential results in an increase in the reaction rate at the electrode and therefore an increased measured current density. In Zone II, a well defined plateau is observed. The reaction rate at the electrode occurs so rapidly that the concentration of the reacting species at the surface of the electrode is essential zero, i.e. the reaction rate at the electrode is greater than the rate of arrival of the reacting species to the electrode. A further increase in the applied potential does not have any effect on the reaction rate. A concentration gradient is therefore created between the

electrode surface (c = 0) and the bulk solution ($c = c_b$). The measured current is dependent only on the diffusion and migration of the reacting species from the bulk solution to the electrode surface. This is called the limiting current condition, or the limiting diffusion condition. In Zone III, the applied potential exceeds the discharge potential of the solvent, where additional current due to secondary reaction (such as hydrogen evolution) at the electrode surface (usually referred to as the hydrogen overpotential condition).

The limiting current condition defined in Zone II is the region where the electrochemical shear method is applicable. Three variables have an affect on this limiting current density: temperature, concentration of the reacting species in the bulk solution, and the relative velocity of flow near the probe surface. Increasing temperature increases diffusivity of the reacting species, thus increasing the rate of diffusion of the reacting species to the electrode. Similarly, increasing the concentration of the species to the probe surface. Finally, an increase in the relative velocity of the liquid flow over the probe results in a reduced concentration boundary layer near the probe surface, thus reducing the diffusion distance. This leads to increased diffusion rate of the species to the probe surface. The electrochemical shear method uses this relationship between flow velocity and diffusion rate to provide information about shear stress based on the measured current.



Applied Potential

Figure 4-1. Typical current-potential relationship when a potential difference is applied between two electrodes immersed in a stagnant solution of reacting species [150]

4.2.2 Relating Rate of Diffusion, Measured Current, and Shear Stress

When a potential is applied between the anode and the cathode, the transport of species from the bulk solution to the surface of the cathode occurs by three mechanisms: (1) migration due to the applied potential field, (2) migration due to the concentration gradient, (3) migration due to convective flow. Migration of species due to the applied potential occurs because of the attraction of the charged species to the opposite charges of the electrodes (i.e., cations towards the negatively charged cathode and the anions towards the positively charged anode). Migration due to the concentration gradient occurs as a result of the difference in the concentration of the species at the reacting electrode (which is essentially zero under the limiting diffusion condition), compared to its concentration in the bulk solution. This concentration gradient provides a driving

force for the diffusion of the reacting species towards the cathodic electrode. Migration due to convective flow occurs if there is a net bulk flow towards the surface of the electrode.

The steady migration of ionic species *i* to the surface of the electrodes can be represented by the following equation, provided transfer is steady [151]:

$$N_i = c_i v_i = -z_i u_i F c_i \nabla \Phi - D_i \nabla c_i + c_i v_i$$
[4-1]

where the three terms on the right hand side of equation 3-23 correspond, in order, to (1) the migration due to the applied potential field, (2) the migration due to concentration gradient, and (3) the migration due to convective flow, as mentioned above. If we consider the case of a horizontal pipe filled with the reacting ionic species i, where the electrodes are mounted on the pipe wall and the species migration occurs only in the y direction toward the electrode, the above equation can be written as:

$$N_{i} = -z_{i}u_{i}Fc_{i}\frac{\partial\Phi}{\partial y} - D_{i}\frac{\partial c_{i}}{\partial y} + c_{i}v_{y} \qquad [4-2]$$

In a scenario where there is no net bulk flow in the y direction, the third term on the right hand side of Equation 4-2 can be considered to be zero. Moreover, if an inert electrolytic solution such as KCl, NaOH, or KOH is added in excess (compared to the reacting species), the overall conductivity of the solution is increased (so that the strength of the electric field becomes negligible), and the potential gradient at the electrode surface, $\frac{\partial \Phi}{\partial y}$ becomes negligible. The migration of species due to the applied potential field can therefore be neglected when compared to the diffusion of the ionic species driven by a concentration gradient [151]. Equation 4-2 can be reduced to:

$$N_i = k \left(c_i - c_b \right) \tag{4-3}$$

Fluctuations in the mass transfer of the species to the surface of the electrodes are directly related to fluctuations in current measured through Faraday's law:

$$N_i = \frac{I}{Av_e F}$$
[4-4]

Therefore combining Equations 4-3 and 4-4 yields the following relationship between mass transfer coefficient and current fluctuations at the electrode:

$$\frac{I}{Av_eF} = k(c_i - c_b)$$
[4-5]

Consider the general convective diffusion equation for flow near the surface of the electrode (in the viscous sublayer of flow regime), which can be represented as

$$\rho\left(\frac{\partial c_i}{\partial t} + v_x \frac{\partial c_i}{\partial x} + v_y \frac{\partial c_i}{\partial y} + v_z \frac{\partial c_i}{\partial z}\right) = \rho D\left(\frac{\partial^2 c_i}{\partial x^2} + \frac{\partial^2 c_i}{\partial y^2} + \frac{\partial^2 c_i}{\partial z^2}\right) + r_i \qquad [4-6]$$

Under steady-state conditions (i.e. assuming that shear or velocity fluctuations are slow compared to the reaction rate at the electrode surface), no chemical generation, diffusion only in the y direction, and flow only along the x-axis, Equation 3-28 can be reduced to

$$v_x \frac{\partial c_i}{\partial x} = D \frac{\partial^2 c_i}{\partial y^2}$$
[4-7]

Moreover, if the Schmidt number is large, i.e. 1000, the mass transfer boundary layer will be very small (which is in the order of 10^{-5} m)¹. As such, it can be assumed that the mass transfer boundary layer lies in the viscous sublayer regime so that du_x/dy is linear. In this range shear stress acting on wall surface is related to the velocity as:

$$v_x = \frac{\tau}{\mu} y$$
 [4-8]

¹ Schmidt number is a dimensionless group that gives an idea of the relative thicknesses of the hydrodynamic boundary layer to the mass transfer boundary layer. The value is the quotient of the absolute viscosity divided by the product of the density and the diffusion coefficient. In mathematical terms, where μ is the absolute viscosity (g/cm-s), ρ is the density (g/cm³), and D is the diffusion coefficient (cm²/s). Large Schmidt numbers as found in liquids are associated with mass transfer boundary layers that are much thinner than hydrodynamic boundary layers.

Equation 4-7 then becomes:

$$\frac{\tau}{\mu} y \frac{\partial c_i}{\partial x} = D \frac{\partial^2 c_i}{\partial y^2}$$
[4-9]

The boundary conditions necessary to solve Equation 4-9 are at y = 0, $c_i = 0$, at $y = \infty$, $c = c_b$ and at x = 0, $c = c_b$. Solving equation 4-9 yields [145]:

$$k = 0.862D \left(\frac{\tau}{\mu D \ d}\right)^{\frac{1}{3}}$$
[4-10]

1

where

$$I = v_e F \frac{\pi d^2}{4} c_b k \qquad [4-11]$$

Figure 4-2 shows the concentration fields and velocity profile over the single probe in a solution of ferricyanide and ferrocyanide as the reversible reacting species.



Figure 4-2. The concentration fields and velocity profile over the single probe [152]

4.2.3 Limitation of the Electrochemical Shear Measurements

There are several limitations to the electrochemical shear method. Firstly, the method is limited to liquids with high Schmidt number (i.e 1000). A high Schmidt number ensures that the hydrodynamic boundary layer near the probe surface is thicker than the mass transfer boundary layer. Secondly, only certain liquids which exhibit the diffusion limiting current conditions can be used (discussed in the next section). Thirdly, the method is limited to probes of a small dimension, since current measurement is related to the spatially averaged mass transfer rate on the surface of the probe. As such, the probe has to be small enough to reflect the shear stress fluctuations near the probe [153]. Finally, the method is limited to systems where the frequency of velocity fluctuations is less than the response time of reaction at the probe surface.

The frequency response of the probes have been studied by several researchers [154-158]. Different models were developed to describe the validity of the electrochemical shear method under fluctuating flow conditions. Sobolik et al. [159] suggested that the method is valid for flow fluctuations less than 100 Hz. Pallares and Grau [153] defined a non-dimensional frequency f^* as :

$$f^* = \frac{2f\delta}{\mu \left(\frac{\tau_w}{\rho}\right)^{1/2}}$$
[4-12]

They suggested that, in a confined channel, the electrochemical method is valid when $f^* < 1$.

4.2.4 Choice of Electrolyte and Electrode

The choice of electrolyte used in the electrochemical shear measurements must meet the following criteria: 1) chemical stability, 2) high solubility, 3) electrode potential different fom that of hydrogen, and 4) low cost [160]. Two popular electrolytic pairs have been used: (1) deposition of copper from copper sulfate solution, and (2) reduction of ferricyanide to ferrocyanide [161]:

(1) $Cu^{2+} + 2e \rightarrow Cu$

(2)
$$\operatorname{Fe}(CN)_6^{3-} + e \rightarrow \operatorname{Fe}(CN)_6^{4-}$$

Under limiting current conditions, a rough deposition of copper on the electrode has been found to increase the surface area on the electrode. As a result, a well defined plateau in Zone II of Figure 4-1. is not observed. Rather, a slow increase in cell current density with increasing applied potential is observed [160]. This makes it difficult to define a range of applied potentials in which the experiments under the limiting current condition can be performed. The reduction of ferricyanide and ferrocyanide does not have this problem, and the plateau seen in Zone II of Figure 4-1is better defined, compared to deposition of copper.

For the ferri-ferrocyanide pair, commonly used electrodes are either nickel or platinum, since both metals are sensitive to the presence of cyanide ions [160]. Moreover, to ensure that rate of reaction at the cathode (the working electrode) is not limited by the rate of reaction at the anode (the reference electrode), two strategies are suggested by Selman [160]: 1) the surface area of the cathode should be much smaller than the surface area of the anode, or 2) the concentration of reacting species at the reference electrode is much higher than the concentration of the reacting species at the working electrode.

4.2.5 Determination of the Concentration of the Inert Salt Solution

As discussed above, one of the assumptions inherent in relating measured current to shear stress is the elimination of migration effect due to the applied potential field. Inert salt solutions such as NaOH, KCl are normally added in excess to reduce/eliminate the migration effect. The following equation can be used to determine the concentration of the salt solution required for a 1% migration current relative to the total migration current [150]:

$$I_{mig} = \frac{1}{2} \frac{C_1^B}{C_{salt}^B} I_1$$
 [4-13]

For example, for a 0.01M ferri and ferrocyanide solution, a concentration of 0.5 M of salt solution is sufficient to reduce the migration current to 1% of total the current [150].

4.2.6 Special Considerations during Experimentation

Special considerations are needed when preparing the electrolyte solution and during experimentation [150], as discussed below.

i. Photochemical Decomposition

Ferricyanide and ferrocynide are sensitive to light, and will undergo photochemical decomposition when exposed to light:

$$Fe(CN)_{6}^{4-} + H_{2}O \xrightarrow{light} Fe(CN)_{5}^{3-} + CN^{-} + H_{2}O$$

$$CN^{-} + H_{2}O \xrightarrow{light} HCN + OH^{-}$$

The decomposition byproduct, hydrogen cyanide (HCN), may poison the electrode surface [150]. As such it is recommended that the solution is not exposed to light, and the solution is stored overnight in an opaque container.

ii. Exposure to Air

Ferricyanide and ferrocyanide are strong oxidizers and react chemically with oxygen when exposed to air. Similar to the byproduct of the photochemical decomposition reactions, the oxidized ferricyanide and ferrocyanide byproducts may also poison the electrode surface [150]. Moreover, when the electrode (platinum) is exposed to air, an oxide film may form on the electrode surface. This may lead to partial or complete blockage of the electrode surface, thus reducing the available surface area for reaction. Isolation of the electrochemical system from air is recommended throughout the experiment.

iii. Effect of Temperature

Diffusivity of ferricyanide is sensitive to temperature changes. Berger and Ziai [150] showed that a 3 °C temperature increase can result in a 20% increase in the limiting current of the system. As such it is important that temperature throughout the experiment be homogenously distributed and constant.

4.2.7 Use of the Electrochemical Shear Probe for Measuring Shear Stress in Gas Sparged Membrane Modules

Recently, with the realization of the importance of shear stress (near membrane surfaces) in controlling fouling, a small number of researchers have started to use electrochemical probes to measure the wall shear stresses on membrane surfaces during filtration. Ducom et al. [93] used an electrochemical probe to measure the shear stress on flat sheet membranes under nitrogen gas bubbling for nanofiltration, and observed a relationship between the time-averaged magnitude and amplitude of the wall shear stress to flux enhancement. Legentilhomme and colleagues [162-165] used an electrochemical probe to study the effect of permeate suction on shear stress, as well as effect of shear stress on cake thickness in a plane ceramic membrane. They observed that suction has an effect on Laborie and Cabassud [100] measured the shear stress for a slug flow shear stress . inside a capillary with the same dimensions as a hollow fiber (inner diameter of 1×10^{-3} m), and speculated that inside the capillary, the liquid film thickness (and hence mass transfer boundary layer) may be controlled more by the solid-liquid interfacial tension rather than hydrodynamics. The experiments conducted by Ducom et al. and Laborie and Cabassud were conducted on rigid surfaces. Recently, Bérubé et al. [107] were the first group to measure the shear stress on the wall of an unconfined non-rigid hollow fiber submerged in a bubbling reactor.

All three studies discussed above regarding the measurement of wall shear stress on membranes, utilized a single probe system, where only the absolute value of the wall shear stress was measured. In a gas-slug two-phase flow membrane system, it may be important to characterize the direction and magnitude of shear caused by the upward liquid slug, the downward falling film around the gas slug and the chaotic turbulent wake at the tail end of the gas slug, all of which may contribute to the removal of foulant or prevention of particle deposit, although the exact contribution of each shear event is not known. This is especially important in an unconfined non-rigid system such as the submerged hollow fiber, where the upward flow characteristics of the rising bubble through the fiber bundle are not known. To date, no literature exists on the characterization of the directional shear stress in a submerged hollow fiber membrane. This is a knowledge gap that impedes the understanding of the mechanisms of permeate flux enhancement in a submerged membrane bioreactors under aeration.

4.2.8 The Double Probe System for Measurement of Directional Shear Stress

The original probe developed by Reiss and Hanratty [145] was capable of measuring the absolute value of the shear stress on the wall, but not the direction and sign of the acting shear stress. As noted in the discussion on gas slug flows above, the sign or direction of the shear stress changes from positive to negative during the passage of the liquid and gas slug as the result of the falling film around the nose of the gas slug. Son and Hanratty [166] and Cognet et al. [167] modified the single probe system, and developed a double-probe system capable of measuring the sign (i.e. direction) of the shear stress on a wall. Since then, the double-probe system has been used extensively by many researchers to study the characteristics of gas-liquid two phase flows [101, 146-148, 152, 168].

The double probe system is constructed using two single probes (probe 1 and probe 2) placed side by side, separated by a very small distance (<0.05 mm) [166, 167], each with its own independent circuit. Since the diameter of the probes is small, the hydrodynamic conditions at the surface of the two probes are assumed to be identical. As such, the signals measured by each probe when operated independently should be identical. When both probes are operated simultaneously (i.e. simultaneous measurement of signal), and

the direction of flow is from probe 1 to probe 2, probe 2 will measure a smaller signal compared to probe 1, since the concentration boundary layer for probe 2 lies in the diffusional wake of probe 1. The magnitude of the shear stress can be calculated form the probe 1 signal, which is unaffected by probe 2. When flow changes direction from probe 2 to probe 1, probe 1 will measure a smaller signal. A comparison between the two signals when probes 1 and 2 are operated simultaneously will therefore provide an indication of the direction of flow. Figure 4-3 shows the concentration fields and the velocity profile over the double probe.



Figure 4-3. The concentration fields and velocity profile over the double probe, when flow is in the direction of probe 1 to probe 2 [152]

4.2.9 Effect of Suction on Wall Shear Stress Measurements

All shear stress measurements mentioned above were conducted without permeation at the membrane wall, i.e. no suction effects. Yucel and Torgoglu [169] postulated that suction at the membrane wall has negligible effect on the tangential velocity profile near the membrane surface, since the suction permeate velocity is very small compared to the axial velocity tangent to the membrane. However, Sofialidis and Primos [170] studied numerically the effect of wall suction on fluid flow and heat transfer at the surface of a porous pipe. The two suction velocities studied (ratio of suction velocity to bulk inlet velocity) were 0.46 % and 2.53 %. It was found that the thickness of the hydrodynamic boundary layer at the pipe surface is reduced due to suction effects. Gaucher et al. [162] and Velikovska et al. [171] measured shear stress at the wall of a plane ceramic membrane during permeation and found that during applied pressure of 50 kPa across the membrane, wall shear stress was increased by approximately 3 to 8 times, and turbulent intensity at the wall due to two-phase flow conditions was dampened or reduced. Their experimental observations were consistent with the findings of Sofialidi and Primos [170], since an increased measured wall shear stress during suction is due to a decrease in the hydrodynamic boundary layer, thereby enhancing the shear stress at the wall.

4.3 Material and Methods

Two different sets of experiments were conducted to validate the shear probes developed. In the first set of experiments, referred to as the Tubular Pipe Wall Experiment, a directional shear probe was fabricated on the inner wall of a vertical tubular pipe, and the directional shear stresses acting on the pipe wall during single-phase and two phase flow conditions were measured. To ensure that the probe was capable of accurate and precise measurement of directional shear stresses, results obtained were compared with theoretically calculated shear stress values (for single-phase pipe flow) and CFD-generated shear stress profiles found in the literature (for two phase pipe flow) for different hydrodynamic conditions.

In the second set of experiments, referred to as the Test-fiber Experiments, a directional shear probe was fabricated on the outer wall of a test-fiber. The test-fiber was a Teflon tube with the same dimensions and flexibility as that of a hollow fiber membrane. The test-fiber was placed at the inner surface of a larger tubular pipe. Shear stress acting on the outer wall of the test-fiber, during single-phase and two phase flow conditions inside the tubular pipe, was then measured. Shear stress profiles obtained in this experiment were compared qualitatively to the shear stress profiles observed in the first set of experiments, as well as to theoretically calculated values for different hydrodynamic

conditions. This set of experiments was conducted to ensure that the probe fabricated on the test-fiber was also capable of precise measurement of directional shear stress.

4.3.1 Tubular Pipe Wall Experiment

Overall System Components

The tubular pipe wall experimental apparatus consisted of a flow cell (which contained the directional shear probe), two vertical pipe segments, a flow recirculation system, a gas sparging system, and an electrical circuit. The schematic diagram of the apparatus is shown in Figure 4-4. The entire apparatus was placed inside a metal wire mesh cage (Faraday cage) which minimized electromagnetic interferences during measurements.



Figure 4-4. The schematic diagram of the experimental apparatus

The flow cell joined the two segments of vertical pipe, which had the same internal diameter as the flow cell (9.9 mm). The length of each pipe segment was 1 m. This length ensured that the entrance and exit effects in flow had negligible effects on shear

measurements at the probe surface, in the range of flow conditions considered. Several experiments were also conducted to check the possible effects of secondary flow generated when the flow cell was not fitted snugly to the pipes. For the flow conditions studied (up to Re = 600) it was found that there was only a negligible effect of this secondary flow on the shear stresses measured.

The directional shear probe consists of two independent shear probes placed next to each other. Two shear probes were fabricated from platinum wire and were mounted onto the inner wall of the flow cell. The surface of each probe was circular with a radius of 0.25 mm. Before mounting into the flow cell, the two probes (referred to as probe 1 and probe 2) were placed side by side, and the gap between them was filled with epoxy to provide probe spacing of less than approximately 0.05 mm. The probes were then inserted into a 1 mm hole in the flow cell, filled with epoxy, and sanded down such that the probes were flush to the inner surface of the flow cell. A schematic diagram of the flow cell is shown in Figure 4-5.



Figure 4-5. The flow cell with the shear probe, used for measuring shear stresses acting on tubular pipe wall

After the mounting process, the probes were examined under a microscope, and the diameter of the probes was measured using the Motic Images (V2.0) software. The software was first calibrated using a calibration slide containing a circle of known diameter. Observations under the microscope, and measurements of the probe diameters indicated that following the sanding process, the final radius of the probes was greater than the original platinum wire radius of 0.25 mm. The measured radius for probe 1 and probe 2 in the flow cell were both 0.27 mm. Additionally, lines on probe surfaces were observed under the microscope, as shown in Figure 4-6. These lines were created during the sanding process, which caused corrugated uneven surfaces on the probes. These uneven surfaces yielded a greater surface available for electrolytic reaction compared to the surface available on a smooth circular surface with a radius of 0.25 mm. To account for this additional surface area, a geometric correction factor was subsequently applied when calculating shear stresses from the measured surface shear signals. The procedure for applying a geometric correction factor is described in Appendix A.



Figure 4-6. Image of the probes mounted on the flow cell, taken using the microscope

The flow recirculation system consisted of a Masterflex pump, a flow rotameter, a gas/liquid separator, and a temperature control bath (Figure 4-4). The gas/liquid separator was immersed in the temperature control bath to maintain the temperature of the electrolyte at 20 °C. The gas sparging system consisted of compressed nitrogen gas and a gas flow meter. Nitrogen gas, instead of air, was used for the two phase flow

experiments because the recirculated electrolyte in the system could react chemically with oxygen, as discussed in Section 4.2.6. The entire flow recirculation system was a closed system to minimized gas leakage and exposure to oxygen.

Electrical Circuit

The cathode of this electrochemical system was the shear probes, and the anode was a piece of stainless steel fitting located on one of the vertical pipes (Figure 4-4). The surface area of the stainless steel pipe fitting was much larger than the surface area of the probes, such that diffusion limiting conditions were only experienced at the probe surfaces.

Each probe had its own independent electrical circuit. An adjustable power supply maintained a consistent differential potential of 300 mV in the circuit. The current in each circuit passed through a 100 ohm resistor, the voltage drop across the resistor was measured, amplified (gain = 1000), and conditioned through a 50 Hz low pass filter (3 - pole butterworth). A data acquisition system with two different channels acquired the signals (i.e. amplified and conditioned voltage drop across resistor) from each of the probes, and recorded real time using a custom Labview application. The data acquisition rate was adjustable. A 1000 Hz acquisition rate was used for all preliminary tubular pipe wall and test fiber experiments. The diagram of the electrical set up is presented in

Figure 4-7.

Experimental Conditions Investigated

Both single-phase and two phase flow conditions were considered. For the single-phase flow condition, liquid flow rates of 0.1 L/min, 0.2 L/min, 0.3 L/min, 0.4 L/min and 0.5 L/min were used. These liquid flow rates were comparable to those in the gas sparged submerged hollow fiber membrane modules [172]. For the two phase flow condition, gas flow rates between 10 and 50 mL/min were used in combination with either 0.2 L/min of liquid flow or zero liquid flow (i.e. gas rising in stagnant liquid).



Figure 4-7. The electrical set-up of the experiment

(1) anode – stainless steel pipe fitting, (2) cathode – shear probes, (3) resistor, (4) 1K amplifier, (5) low pass filter, (6) data acquisition box, (7) computer for data logging

4.3.2 Test-fiber Experiment

Overall System Components

The test-fiber experimental apparatus was similar to that of the tubular pipe wall apparatus. The same flow recirculation system, electrical circuit and gas sparging system was used for the two sets of experiments. The flow cell and the pipe segments, however, were different. The flow cell in the test-fiber experiment consisted of a test-fiber on which a probe was mounted, and a flow cell that held the test-fiber in place inside a tubular pipe. The section below describes in detail the test-fiber, the flow cell and pipe segment used in the test-fiber shear experiment.

Test-fiber

A hollow Teflon tube with similar dimensions and flexibility to those of a hollow fiber membrane (o.d. 1.7 mm) was used. This Teflon tubing used as a surrogate for a hollow fiber membrane is hereafter referred to as a "test-fiber". Two shear probes were mounted onto the surface of the test-fiber. Similar to the probe mounted on the tubular pipe flow cell, the surface of each probe on the test-fiber was circular with a radius of 0.25 mm, fabricated from platinum wire. Since the two platinum wires were required to be placed side by side, it was difficult to fit the two wires into the cavity of the test-fiber (i.d. 0.9 mm). As such, a mold with an identical outer diameter to that of the test-fiber was created, such that the two platinum wires could be fitted inside the cavity of the mold, and the mold was then filled with epoxy. The total length of the mold was approximately 1 cm. Both ends of the mold containing the platinum wire were joined to the test-fiber to make a continuous fiber length. A picture of the double probe on the mold is shown in Figure 4-8.



Figure 4-8. A picture of the shear probes on the test-fiber a) top view, b) side view

The measured projected radii of both probes after the mounting process were both 0.285 mm (see Appendix B). Unlike the probes described in the Tubular Pipe Wall section (Section 4.3.1), no geometric calibration was applied for the probes mounted on the test-fiber, since the flow cell geometry in which the test-fiber was placed was not a perfectly

circular pipe wall, rather, the test-fiber protruded slightly into the pipe wall. Instead, the signal of probe 2 was normalized against the diameter of probe 1 (such that both probes measured the same signal under the same flow conditions.)

Flow Cell and Vertical Tubular Pipe

A support block with a groove (0.7 mm wide and 8 cm long) machined onto the surface was created. The section of the test-fiber with the shear probe (facing outward) was fitted snuggly onto the groove. A 0.7 mm wide and 6 cm long slot was machined onto a tubular pipe (i.d. 9.9 mm), and the rectangular block with the test-fiber was pressed against the slot of the pipe such that the probes on the test-fiber faced in the inner walls of the pipe. A schematic diagram of this set-up is shown in Figure 4-9.





(a) overall system, (b) cross section A-A

Experimental Conditions Investigated

Both single-phase and two phase flow conditions acting on the test-fiber inside the tubular pipe were considered. For the single-phase flow condition, liquid flow rates of 0.1 L/min, 0.2 L/min, 0.3 L/min, 0.4 L/min and 0.5 L/min were used. For the two phase flow conditions, gas flow rates between 10 and 50 mL/min were used in combination with either 0.2 L/min of liquid flow or zero liquid flow (i.e. gas rising in stagnant liquid).

4.3.3 Electrolyte

A search in the literature revealed that the pairing of ferricyanide (0.003 M) with ferrocyanide (0.006 M) had been successfully applied in electrochemical shear measurements [173]. Potassium chloride (0.3M) was used as the supporting electrolyte. The experiments described in this section were conducted using this electrolyte composition, with deionized water. The physical properties of these electrolytes are shown in Table 4-1.

Compound	Concentration	Molecular weight	Amount
Potassium ferricyanide	0.003 M	329.26	0.988 g
Potassium ferrocyanide	0.006 M	422.41	2.534g
Potassium chloride	0.3 M	74.56	24.60 g
Deionized water			1 L

Table 4-1. Physical properties of the electrolytes

Physical properties of electrolyte at 20°C [173]

Density =1016 kg/m3

Viscosity = 0.001kg/m/s Ferricyanide diffusivity = 7.14×10^{-10} m²/s

4.3.4 Shear Stress Measurement

A detailed description of the procedure for measuring shear stress using the probes is described in Appendix C. Prior to the start of each experiment, the entire apparatus was purged with nitrogen gas. The surface of the anode and probes were also cleaned with Q-

tips and de-ionized water after each set of experiments (i.e. often approximately 5 minutes).

Magnitude of Shear Stress

When measuring shear stress using probe 1, probe 2 was unplugged from the circuit so that it would not interfere with the measurements from probe 1 (and vice versa). Measured signals for the probe were calibrated geometrically (described in Appendix A). Signals from probes 1 and probes 2 were also corrected by accounting for the intrinsic electrical instrument bias (discussed in Section 4.3.6).

As previously discussed, the current through the shear probe (i.e. cathode) could be converted to a surface shear stress measurement. In the present study, the current was measured based on the voltage drop through a resistor (Figure 4-7). As a result, the following relationship (which was derived from equations 4-10 and 4-11) was used to calculate the shear stress acting on the probe surface based on the corrected voltage reading (V_{cal}), which took into consideration the geometric calibration factor and the instrument bias (Appendix A and D):

$$\tau = 99.92 \left[\frac{V_{cal}}{\pi v F C R G} \right]^3 \frac{\mu}{d^5 D^2}$$
 [4-14]

Direction of Shear Stress

Indication of flow direction can be achieved by simultaneously measuring the current from both probes mounted on the wall surface during liquid flow through the tubular pipe. When two probes are placed in close proximity, the signals obtained from both probes should be similar. However, when the direction of flow is from probe 1 to probe 2, probe 2 will measure a smaller signal compared to probe 1, since the concentration boundary layer of probe 2 lies in the diffusional wake of probe 1 (as discussed in Section 4.2.8). In this case, the true signal is obtained from probe 1, and probe 2 provides information on the direction of the flow. The opposite occurs when the direction of flow is from probe 2 to probe 1 (Figure 4-10).



Figure 4-10. The use of double probe system for measurement of directional shear stresses

(a) direction of flow from probe 1 to probe 2, (b) direction of flow from probe 2 to probe 1

4.3.5 Sources of Error in Shear Measurement

Potential sources of error in calculating shear stress from the measured surface shear signal are associated with errors in measuring (or estimating) the following parameters in Equation 4-14:

- probe geometry (i.e. available surface area for reaction at cathode),
- ferricyanide diffusivity (temperature dependent),
- ferricyanide concentration,
- electrolyte density (temperature dependent),
- electrolyte viscosity (temperature dependent), and
- liquid flow rate.

A sensitivity analysis was conducted to establish the effect of changes in the parameters in Equation 4-14. The sensitivity analysis was based on the effect of the percent change of the parameters, and the resulting effect on the percent change in the calculated shear stress. Figure 4-11 shows the sensitivity of the calculated shear stress value for each of these variables, measured in terms of the percent change of the variable and its effect on the percent change in the calculated shear stress. As illustrated, small changes in the probe diameter have the highest effect on the calculated shear stress. As discussed previously, it was not easy to control the diameter and surface of the probe during the probe fabrication process. Therefore, the measured signals were corrected with a geometric correction factor to account for the fact that the effective diameter of the probe varied slightly (Appendix A).



Figure 4-11. Sensitivity analysis of the calculated shear stress value for different parameters, measured in terms of the percent change of the variable and the resulting effect on the percent change in the calculated shear stress

4.3.6 Instrument Bias and Signal Noise

The data acquisition system consisted of 2 channels available for acquiring surface shear signals. Each channel had an intrinsic instrument bias, where a small signal was measured even when the data acquisition system was isolated from the electric circuit. Measured surface shear signals from all experiments were corrected for these instrument biases, as described in detail in Appendix D.

4.3.7 Limiting Current Test

An experiment was performed to check the limiting current of the electrochemical system. The experiments were conducted using the pipewall experiment set-up, with varying liquid flow rates. The current through the circuit was measured for different applied potentials. The limiting current or diffusion condition was achieved when applying a potential in the range of 200 mV to 400 mV. The limiting current test is described in Appendix E. For the experiments described in the rest of the chapter, a potential of 300 mV was used.

4.3.8 **Re-use of Electrolytes**

As discussed in Chapter 3, possible photochemical decomposition and oxidation reactions of ferricyanide and ferrocyanide may degrade the quality of the electrolyte. An experiment was conducted in three different days using the same electrolyte to check if the possible degradation of the electrolyte had an effect on shear measurements. Careful cleaning of the shear probe and the stainless steel pipe fitting was performed prior to each experiment. It was found that with careful cleaning of the probes and anode before each experiment, effects of electrode poisoning due to photochemical decomposition and oxidation reactions could be avoided, and the reaction byproducts in the electrolyte solution did not have a substantial effect on shear measurements (see Appendix F). For the present study, electrolyte were discarded every three days, and a new batch of electrolyte was created for subsequent experiments.

4.4 Results

4.4.1 Tubular Pipe Wall Shear Measurement

Single-phase Flow Conditions

The measured surface shear stresses were compared with theoretically calculated shear stresses (see Appendix A) for single-phase pipe flow for flows between 0.1 and 0.5 L/min, as shown in Table 4-2 and Table 4-3. The error of the measured shear stress, after the geometric correction, ranged from 1% to 20% for both probes. Although the application of the geometric correction factor was an approximated approach, the values shown in Table 4-2 and Table 4-3 show that the probes developed indicated shear stresses were similar to the theoretically calculated shear stresses, for the range of flow rates investigated.

Liquid Flow Rate (L/min)	Measured Shear Stress (Pa) * ²	Theoretical Shear Stress (Pa)	Difference between Measured and Theoretical Shear Stress (%)
0.1	0.021	0.018	20 ± 1
0.2	0.041	0.035	17 ± 1
0.3	0.052	0.053	1 ± 1
0.4	0.068	0.070	2 ± 1
0.5	0.085	0.088	3 ± 1

Table 4-2. Comparison between measured and theoretically calculated shear stress for Probe 1

* Corrected for instrument bias, and based on geometric correction factor of 1.03

 $^{^{2}}$ The standard deviation of the measured shear stress is not shown, however, the standard deviation of the measured shear signal ranged from 0.005 to 0.008 V.

Liquid Flow Rate (L/min)	Measured Shear Stress (Pa) * ²	Theoretical Shear Stress (Pa)	Difference between Measured and Theoretical Shear Stress (%)
0.1	0.020	0.018	13 ± 0.01
0.2	0.035	0.035	1 ± 0.01
0.3	0.048	0.053	$8~\pm~0.01$
0.4	0.060	0.070	$14\pm\ 0.01$
0.5	0.072	0.088	$18\pm\ 0.01$

Table 4-3. Comparison between measured and theoretically calculated shear stress for Probe 2

* Corrected for instrument bias, and based on geometric correction factor of 1.06

The probes were also observed to be capable of indicating the direction of the shear stress, when the direction of flow was changed. When the flow was from probe 1 to probe 2, probe 1 measured a higher signal. When the flow was from probe 2 to probe 1, probe 2 measured a higher signal. Also, increasing the flow rate in the pipe resulted in an increase in the difference between the signals measured from probes 1 and 2 (Appendix G). This observation can be attributed to the fact that an increased flow rate resulted in a more significant diffusional wake behind the upstream probe.

Two Phase Flow Conditions

In this section, the wall shear profiles of two different types of two phase flow condition in the vertical pipe are presented: 1) gas flow in moving liquid, and 2) gas flow in stagnant liquid. Figure 4-12 shows the raw surface shear signals (V_{cal}) recorded from Probe 1 and Probe 2 induced by a series of gas slugs rising in moving liquid. V_{cal} corresponds to the amplified and conditioned voltage drop measured across the resistor (

Figure 4-7), and the signal was also corrected for instrument bias and geometric correction factor. The gas flow rate was 50 mL/min (superficial velocity $v_g = 0.01$ m/s), and the liquid flow rate was 0.2 mL/min (superficial velocity $v_L = 0.04$ m/s).


Figure 4-12. Typical V_{cal} signals recorded from Probe 1 and Probe 2 induced by a series of gas slugs rising in moving liquid

Gas slug rising in the direction from Probe 1 to Probe 2. Liquid flow rate = 0.2 L/min, gas flow rate= 50 mL/min

The reconstructed wall shear stress profile, calculated based on V_{cal} using Equation 4-14, is shown in Figure 4-13a. A magnified view of the shear profile during the passage of a single gas slug (at 46 seconds) is presented in Figure 4-13b. This shear profile is similar to that obtained experimentally by others during slug flow in a vertical pipe (Figure 3-4), where positive shear stress was observed during the passage of a liquid slug, and negative shear stresses were observed during the passage of the gas slug [97, 101, 148, 174, 175]. The measured shear stress during the passage of a gas slug was higher than that during the passage of the liquid slug. Although the gas and liquid superficial velocities were different than that for the experimental results presented in Figure 3-4, the magnitude of shear stresses in Figure 4-13 are similar to those of Figure 3-4 (shear ranges from 0.2 Pa for liquid slugs to -1.4 Pa for gas slugs).

Figure 4-14 shows the shear profile of a single gas slug (length 11 cm, volume = 8.5 mL) rising in the stagnant liquid in a vertical pipe. Similar to the profile in Figure 4-13b, shear stress was negative during the passage of the gas slug. Since there was no liquid slug, the shear stress before and after the passage of the gas slug was zero. The shear



profile seen in Figure 4-14 is very similar to that obtained computationally using CFD by others for a slug rising in stagnant liquid in a vertical pipe(Figure 3-3).





(b)

Figure 4-13. Typical wall shear stress profile of gas slugs rising in moving liquid (calculated based on the raw surface shear signals from Figure 4-12)

(a) Data over 30 seconds, (b) Data over 0.65 seconds at 46th second



Figure 4-14. The shear profile of an 11 cm long gas slug (8.5 mL) rising in stationary liquid in a vertical pipe (i.d. 9.9 mm)

4.4.2 Test-fiber Surface Shear Measurement

Single-phase Flow Condition

The measured surface shear stresses on the test-fiber were compared with theoretically calculated shear stresses (see Appendix A) for single-phase pipe flow for flows between 0.1 and 0.5 L/min, as shown in Table 4-4. The error of the measured shear stress, after the geometric correction, ranged from 12% to 64% for both probes. However, the comparison of the theoretical and measured shear stresses using the test-fiber is not ideal, for the following reasons:

(1) the shear probe diameters of the test-fiber were estimated, and

(2) theoretically calculated shear stress based on shear acting on smooth tubular pipe wall, while the surface on the flow cell which houses the test-fiber was not smooth ³.

As such, differences between the two values were expected. However, the comparison between the two shear stresses shows that the experimentally observed shear stress values were of the *same order of magnitude* as the theoretically calculated shear stresses. This indicates that the test-fiber could be used as a tool to capture trends in shear stress under different hydrodynamic conditions.

Table 4-4. Comparison between measured and theoretically calculated shear stress for Probe 1

Liquid Flow Rate (L/min)	Measured Shear Stress (Pa) *	Theoretical Shear Stress (Pa)	Difference between Measured and Theoretical Shear Stress (%)
0.1	0.028	0.018	61 ± 1
0.2	0.051	0.035	45 ± 1
0.3	0.075	0.053	44 ± 1
0.4	0.096	0.070	37 ± 1
0.5	0.119	0.088	36 ± 1

* Corrected for instrument bias.

 $^{^{3}}$ No numerical solution exists to theoretically calculate wall shear stress for the geometry presented in Figure 4-6.

Two Phase Flow Conditions

The shear stresses acting on the surface of the test-fiber (held in a vertical tube) as gas slugs rose along the test-fiber is presented in

Figure 4-15. The gas slugs delivered were large enough that they assumed the profile of Taylor bubbles. For this set of experiments the liquid flow rate was 0.4 L/min, and the magnitude of the measured signals in the falling film region were approximately twice the magnitude of those presented in Figure 4-12. Possible reasons for this difference are:

- 1. the surfaces surrounding the probes and test-fiber in the flow cell were not identical to those of a smooth tubular pipe wall,
- 2. the placement of the test-fiber in the flow cell tubular pipe resulted in a smaller cross-sectional area available for the rising gas slug, and
- 3. the geometry of the probes on the test-fiber was different than that in the tubular pipe, due to a larger curvature effect on the test-fiber (diameter 1.7 mm) than on the pipe wall flow cell (diameter 9.9 mm). During the mounting process, the probes on the test-fiber were sanded down such that their surfaces were flush with the surface of the test-fiber. Since the surface of the test-fiber was curved, the true probe diameter is greater than the measured diameter (discussed in Appendix B).

Based on the discussions above, there was also no theoretical basis for applying a geometric correction factor based on numerical estimates of the theoretical shear stresses on vertical pipe walls.

Despite this discrepancy between the theoretically calculated and the experimentally measured shear stress, however, the shear profiles measured using the probes on the test-fiber were similar to those seen in Figure 3-4 for the passage of gas slugs. Positive shear stress was observed during the passage of a liquid slug, and negative shear stresses were observed during the passage of gas slugs. The measured shear stresses during the

passage of a gas slug were higher than the shear stresses measured during the passage of the liquid slug. These profiles were reproducible as well. As the discussions presented in Chapters 5 to 8 focus on shear profiles and relative shear values, rather than absolute shear values, these results indicate that the test-fiber can be used as an effective measurement tool for the present study. This suggests that the test-fiber can be used as a tool to capture trends in shear stress under different flow conditions. This is sufficient for the subsequent investigations reported in Chapters 5 to 8.







Figure 4-15. The shear stresses acting on the surface of the test-fiber (held in a vertical tube) as three gas slugs rises along the test-fiber

Raw shear Raw surface shear signals recorded from Probe 1 and Probe 2, (b) calculated shear stress. Liquid flow rate 0.4 L/min

4.5 Conclusions

Electrochemical shear probes were developed to measure the magnitude and direction of shear stress under single and two phase flow conditions. Two types of shear probes were developed, one for measuring shear stress acting on the inner wall of a vertical pipe, and one for measuring shear stress acting on the outside surface of a test-fiber placed inside a vertical tubular pipe. Geometric correction factors were applied to the probes in the vertical tubular pipe, but not the probes on the test-fiber. The following are the conclusions from these studies:

- The two probe system developed was capable of capturing the shear profiles generated under single and two phase flow conditions. For the range of singlephase flow rates examined (0.1 to 0.5 L/min), the measured surface shear signals were comparable to theoretically calculated shear stresses.
- 2. In the tubular pipe experiment, the deviation from theoretically calculated shear stress ranged from 1 to 20%. In the test-fiber experiment, the deviation from theoretically calculated shear stress ranged from 12 to 64%. Although the error was larger for the test-fiber experiments, the measured shear stress values were judged to be satisfactory, given the significant differences in the assumptions involved in each approach to estimation of shear stress.
- 3. For two phase flow conditions, the shear profiles of a rising gas slug measured using both types of shear probes on the test-fiber were similar to those observed experimentally and to those generated using CFD. As such, the shear probes developed on the test-fibers were concluded to be satisfactory for capturing trends in shear stress under different hydrodynamic conditions.

5. Shear Profiles Inside Gas-Sparged Submerged Hollow Fiber Membrane Modules

5.1 Introduction

Many strategies have been developed to control and minimize membrane fouling. As discussed in Chapter 3, one such strategy is the control of hydrodynamic conditions near membrane surfaces to prevent particle deposition on the membranes, thereby reducing the extent of membrane fouling and enhancing permeate flux. Gas sparging at the bottom of the membrane units is commonly used to achieve these favorable hydrodynamic conditions. The mechanisms by which gas sparging increases the back-transport of material away from membrane surfaces may depend on the membrane configuration used (i.e. confined vs. unconfined membranes) [112]. Confined modules, such as tubular membranes, are modules in which the feed water and sparged gas bubbles are confined within the membranes in an inside-out permeate flow configuration. Unconfined modules, such as submerged hollow fibers or flat sheet membranes, have an outside-in flow configuration. The extent of fouling reduction in a confined membrane module is dependent on the bubble dynamics inside the confined membrane. Slug flow has been found to minimize fouling on membrane surfaces to a greater extent than bubble, churn and annular mist flows in confined membrane systems [10, 87, 91, 97, 98]. The mechanism of fouling reduction and flux enhancement for slug flow inside a confined membrane system has been linked to the high shear stresses induced by liquid flow reversal in the falling film surrounding a rising gas slug, the oscillating flows near the tail end of the rising gas slug, and the overall increase in the bulk superficial velocity imparted by the rising gas slug [10].

In unconfined systems such as submerged hollow fiber membranes, very limited experimental literature exists on the mechanisms by which fouling reduction and flux enhancement is achieved. When studying the effect of gas sparging on submerged hollow fibers, some researchers have assumed that since the packing density of the fibers

is high, the surface shear stresses induced by gas bubbles rising between fibers are similar to those induced by gas slugs in a confined system. As a result, the mechanisms of flux enhancement in unconfined membrane systems has often been assumed to be similar to those of confined membrane systems [10, 139]. The validity of this assumption is questionable, given the complex and transient nature of the geometry and the hydrodynamic conditions in submerged membrane modules (see Section 3.6). Depending on the aeration system, bubbles with different sizes and shapes can form in an unconfined system (i.e. spherical bubble, ellipsoidal bubble or spherical cap bubbles), and each bubble geometry yields different rise velocities and subsequently results in different shear stresses acting on the membrane surface. Loosely held fibers in the submerged hollow fiber system may sway laterally as a result of the wakes generated by the rising bubbles. The hydrodynamic conditions surrounding gas-sparged submerged hollow fibers are poorly understood. This knowledge gap presents a real impediment to developing a complete model of the filtration process and to optimizing the performance of submerged hollow fiber membranes in terms of membrane geometry and energy costs associated with bubbling.

The objective of the study presented in this chapter was to address the knowledge gap discussed above from the point of view of hydrodynamics. More specifically, the question of whether the hydrodynamic conditions near the membrane surfaces of unconfined systems are similar to those observed in confined systems was addressed. The directional surface shear stresses generated by rising bubbles in tightly and loosely held fiber bundles were measured for different fiber packing densities, diffuser nozzle sizes and gas sparging rates, and compared to those in the confined system.

5.2 Materials and Methods

The experimental system consisted of a test-fiber with a directional shear probe (testfiber), an electrolytic solution, a data acquisition system, fiber bundle, a system tank, a gas sparging system, and a high speed imaging system. The schematic diagram of this set-up is shown in Figure 5-1. The experimental program was based on a factorial-type design which considered different module configurations, fiber packing densities in a bundle, diffuser nozzle sizes and gas sparging rates.



Figure 5-1. The schematic diagram of the bench-scale experimental system

The directional shear probe installed on a test-fiber was described in detail in section 4.3.2. The electrolytic solution was described in detail in Section 4.3.3. The data acquisition system was described in detail in Section 4.3.1. For each experiment, shear measurements were collected for a period of 10 seconds at a rate of 1000 Hz, generating a total of 10,000 data points. Each measured value was referred to as a "frame number" which corresponds to the image number captured using a high speed video camera . In the present study, the shear stress measurements are presented in units of volts, instead of shear stress (i.e. Pa). The shear measurements were corrected for instrument bias. To convert voltage measurements to shear stress, it must be assumed that (1) the axial diffusion is negligible, (2) the velocity gradient in the direction normal to the electrode

surface is negligible, (3) a linear velocity gradient in the region of measurement (in the thin concentration boundary layer) exists [161], and (4) the cathode is circular, and (5) the frequency response of the probe is faster than the rate of low fluctuation near probe surface. Although these assumptions are expected to be valid for the experimental set-up used in the present study, voltage signals are presented, since only shear profiles and relative shear values, rather than absolute shear values, are of interest.

For all experiments, the hollow fibers used consisted of Teflon tubes with similar dimensions and flexibility as those of commercially available hollow fiber membranes (o.d. 1.7 mm). Teflon tubes were used instead of actual hollow fiber membranes because the Teflon tubes are translucent, which enabled images of rising bubbles within the submerged fibers to be captured by the high speed camera. The Teflon tubes are referred to as "fibers" for the rest of the thesis. The length of each fiber in a bundle was 12 cm, and the fibers were held in place at both ends by a circular disk, as presented in Figure 5-2. Three fiber packing densities were studied: low, medium, and high fiber packing densities (8, 16 and 32 fibers/cm², respectively.)

All fibers were arranged in a bundle with a square matrix arrangement. For the bundle with low fiber packing density, 4 fibers, including the test-fiber, were inserted into the holes in the circular disks to form a square matrix. Each fiber was separated by an empty 1.6 mm hole in the circular disk, as shown in Figure 5-2a. The resulting distance between each fiber was 3.6 mm. For the bundle with medium fiber packing density, 8 fibers, including the test-fiber, were inserted into the holes in the circular disks (Figure 5-2b). The distance between fibers was 1mm. For the bundle with high fiber packing density, 8 fibers were placed side by side in a square matrix with no space between each fiber (except for a hole in the middle of the bundle for the diffuser nozzle), and glued together with epoxy at both ends (Figure 5-2c). A circular disk with a large hole drilled in the middle was used to hold this bundle at both ends. In all of the arrangements, the shear probe was positioned at mid length of the test-fiber (i.e. 6 cm from the bottom of the bundle).



Figure 5-2. Top and side views of the fiber arrangements in the bundles with different fiber packing densities

a) low packing density, b) medium packing density, c) high packing density

The fiber bundles were immersed in a cylindrical system tank filled with the electrolyte solution. Both ends of the bundle (i.e. circular discs) were held in place such that the fibers could be held either loosely or tightly by adjusting the height at the top of the bundle. For tightly held fibers, the distance between the circular disks which held the ends of the fibers was 12 cm. For loosely held fibers, the distance between the circular disks which held the circular disks was 95 % of this value (i.e. 11.4 cm).

The cylindrical system tank was constructed of plexiglas so that bubbles in the tank could be observed visually. The system tank was sealed at the top to prevent oxygen exposure during each experiment. A nickel anode was also immersed in the system tank.

Nitrogen gas was delivered to the center of the bundles using diffuser nozzle sizes of 1 mm or 2 mm (i.d.). Nitrogen gas, instead of air, was used because ferricyanide and ferrocyanide reacts chemically with oxygen. Three different gas sparging rates were investigated: 2 mL/min, 10 mL/min and 35 mL/min.

A high speed video camera (Kodak Ektapro 1000 Motion Analyzer System) was used to take images of the sparged bubbles as they rose through the fiber bundles. The high speed camera captured images at a rate of 1000 Hz, which was the same frequency at which the surface shear signals were acquired. The camera was connected to the data acquisition system, so that the sampling of the surface shear signals was synchronized to the video camera image acquisition. A small section of the cylindrical system tank was cut out and replaced by a flat panel made of plexiglas so that images taken by the video camera were not optically distorted by the curvature of the cylindrical tank.

5.2.1 Experimental Program

An experimental program which considered different module configurations (loosely and tightly held), fiber packing densities (8, 16, 31 fibers/cm²), diffuser nozzle sizes (1 mm, 2 mm) and gas sparging rates (2, 10, 35 mL/min) was conducted to quantify shear stresses acting on gas-sparged submerged hollow fiber modules. Each experimental condition was repeated three times, and in random order.

All experiments were conducted at room temperature (23 °C) and under conditions of limiting current. A summary of all experiments performed is provided in Table 5-1. The first number in the notation used to name the different experiments refers to diffuser nozzle size used (i.e. 1 or 2 mm). The subsequent letter and number correspond to the fiber packing density (i.e. A, B or C for low, medium or high fiber packing densities,

respectively) and the gas sparging rate (i.e. 1, 2, or 3 for 2, 10 or 35 mL/min, respectively), respectively.

Experiment	Packing	Nozzle Size	Gas Flow	Fiber Configuration
No.	Density	(mm)	(mL/min)	
1-A1	Low	1	2	Tightly held
1 - B1	Med	1	2	Tightly held
1-C1	High	1	2	Tightly held
2-A1	Low	2	2	Tightly held
2-B1	Med	2	2	Tightly held
2-C1	High	2	2	Tightly held
1-A2	Low	1	10	Tightly held
1-B2	Med	1	10	Tightly held
1-C2	High	1	10	Tightly held
2-A2	Low	2	10	Tightly held
2-B2	Med	2	10	Tightly held
2-C2	High	2	10	Tightly held
1-A3	Low	1	35	Tightly held
1-B3	Med	1	35	Tightly held
1-C3	High	1	35	Tightly held
2-A3	Low	2	35	Tightly held
2-B3	Med	2	35	Tightly held
2-C3	High	2	35	Tightly held
1-A3	Low	1	35	Loosely held
1-B3	Med	1	35	Loosely held
1-C3	High	1	35	Loosely held
2-A3	Low	2	35	Loosely held
2-B3	Med	2	35	Loosely held
2-C3	High	2	35	Loosely held

Table 5-1. Summary of the experimental program

5.3 Results

5.3.1 Bubble-Induced Shear Profile for Tightly Held Fiber Bundles

Typical surface surface shear signals obtained for the tightly held fibers in low, medium and high packing density (L, M,H) bundles at different gas sparging rates (2, 10 and 35 mL/min) and diffuser nozzle sizes of 1 mm and 2 mm are shown in Figure 5-3 and Figure 5-4. As expected, for all experiments, increasing the gas sparging rate increased the "baseline" surface surface shear signals. The baseline surface shear signal obtained for the experiment with a low fiber packing density and a gas sparging rate of 2 mL/min (graph aL, Figure 5-3) was approximately 0.2 V. For the same bundle, increasing the gas sparging rate to 10 and 35 mL/min increased the baseline surface surface shear signals to 0.4 V and 0.5 V, respectively (bL and cL, in Figure 5-3 respectively). The increase in the baseline surface shear signals likely resulted from an increase in the bulk superficial liquid velocity at the higher gas sparging rates. However, the contribution of the bulk superficial liquid velocity to overall surface shear signal is quite small when compared to that induced by rising bubbles. This is consistent with the observations reported by Bérubé et al. [107]. As presented in Figure 5-3 and Figure 5-4, sparged bubbles induced instabilities which generated relatively large peaks in the surface shear signals. The peak surface shear signals induced by the instabilities were typically 2 to 5 times higher than those associated with the baseline surface shear signals.



Figure 5-3. Surface shear signals measured from test-fiber at different fiber packing densities and gas sparging rates for tightly held fibers and 1 mm gas diffuser

a) gas flow 2 mL/min, b) gas flow 10 mL/min, c) gas flow 35 mL/min. Dashed line corresponds to baseline shear signal



Figure 5-4. Surface shear signals measured from test-fiber at different fiber packing densities and gas sparging rates for tightly held fibers and 2 mm gas diffuser

a) gas flow 2 mL/min, b) gas flow 10 mL/min, c) gas flow 35 mL/min. Dashed line corresponds to baseline signals

5.3.1.1 Number of Surface Shear Events, and Magnitude of Surface Shear Signals

In Figure 5-3 and Figure 5-4, each peak in the surface shear signal (with amplitude greater than 0.1 V relative to the baseline and lasting longer than 0.5 seconds) was considered to be a surface shear event caused by the passage of a rising gas bubble. The number of surface surface shear events observed for different fiber packing densities, diffuser nozzle sizes and gas sparging rates for the replicate experiments is summarized in Figure 5-5. The error bars on each point correspond to a confidence interval equivalent to one standard deviation of the measured surface shear events of the replicate experiments. Although the error bars overlap, several repeatable trends were observed for each of the replicate experiments. These trends are discussed in the following section.

For all experiments, increasing the gas sparging rate increased the number of surface shear events. This was expected, since the number of gas bubbles introduced into the system increased as the gas sparging rate was increased. When comparing the number of surface shear events obtained for the three different fiber packing densities, the experiments performed with medium fiber packing density generally resulted in the highest number of surface shear events, compared to those with low and high fiber packing densities. For low fiber packing densities, the rising bubbles were not completely and consistently confined by the fibers, especially at higher gas sparging rates (i.e. 10 and 35 mL/min). As a result, some bubbles were observed to periodically escape from the bundles through the relatively large gaps that existed between the fibers. The non-confining nature of the low fiber packing density bundle lowered the number of surface shear events. This phenomenon can be seen in graph bL of Figure 5-4, where no surface shear events were observed between frames 5000 and 7000 as a result of bubbles escaping the bundle before reaching the shear probe. A different phenomenon occurred to generate relatively low number of surface shear events for bundles with high fiber packing density. Due to the high fiber packing density, the bubble break-off rate at the diffuser nozzle was less than that for the bundles with low and medium fiber packing densities. As a result, fewer and larger bubbles, resembling gas slugs rising in a confined system, were observed for high fiber packing density bundles. Typical images of bubbles rising in medium and high fiber packing density bundles are shown in Figure 5-6b and Figure 5-6c, respectively.

As presented in Figure 5-5, it was observed from Figure 5-5 that the number of shear events appeared to be proportional to flow (for medium packing density). This suggests bubbles of relatively constant size for all flows. Also, for experiments with the low and medium fiber packing densities, the number of surface shear events obtained was consistently higher when sparging with a 1 mm diffuser nozzle when compared to sparging with the 2 mm diffuser nozzle. For experiments with high fiber packing density, however, the opposite trend was observed. The reason for this discrepancy was that bubbles delivered through the 2 mm diffuser nozzle broke apart to form smaller bubbles when they came into contact with the surrounding closely packed fibers. On the other hand, frequent bubble break-up was not observed for bubbles delivered through the 1 mm diffuser nozzles. As a result, fewer and larger bubbles were observed for high fiber packing density bundles. Typical images of the bubbles delivered through the 1 mm and 2 mm diffuser nozzle for high fiber packing density bundles are shown in Figure 5-6b and Figure 5-6c, respectively.



Figure 5-5. The number of surface shear events (over a period of 10 seconds) experienced by tightly held fibers during gas sparging using diffuser nozzles sizes of 1 mm and 2 mm

(a) gas sparging rate 2 mL/min, (b) gas sparging rate 10 mL/min, (c) gas sparging rate 35 mL/min. Note the scale for (a) is different than that of (b) and (c). The error bars on each point correspond to confidence interval equivalent to one standard deviation of the measured surface shear events of replicate experiments

(b)





Figure 5-6. Typical images of bubbles rising in fiber bundles. The translucent long streaks seen in the pictures are the 8 fibers in the bundle. The black dashed line highlights the bubbles rising within the fiber bundle

(a) medium fiber packing density, gas sparging 35 mL/min, diffuser nozzle size 1 mm, tightly held fibers. The bubble was approximately spherical.

(b) high fiber packing density, gas sparging 35 mL/min, diffuser nozzle size 1 mm, tightly held fibers. The bubble delivered through the 1 mm diffuser nozzle is longer than the bubble in Figure 5-6c.

(c) high fiber packing density, gas sparging 35 mL/min, diffuser nozzle size 2 mm, tightly held fibers. The bubble is longer than the bubble in Figure 5-6a.

(d) medium fiber packing density, gas sparging 35 mL/min, diffuser nozzle size 2 mm, loosely held fibers. Bubbles are not confined within the fiber bundle, but are in close proximity to the fibers.

The average surface shear signals for experiments performed at different gas sparging rates and fiber packing densities are presented in Figure 5-7. As shown, increasing the gas sparging rate increased the average surface shear signals. However, increasing the fiber packing density did not have a significant impact on the average shear signal. Also, sparging through 1 mm or 2 mm nozzles did not result in significant differences in the average shear signal.

As discussed in Section 3.7, peak shear stress and the standard deviation of the shear stress have been reported to be linked to fouling. The effect of average, peak and standard deviation shear on fouling control is investigated in Chapter 8.



Figure 5-7. Average surface shear signal for each experiment with different diffuser nozzle sizes

(a) 1 mm nozzle, (b) 2 mm nozzle. Error bars correspond to confidence interval equivalent to one standard deviation of shear in the replicate experiments

Several studies have reported that gas sparging with small bubble diffusers (i.e. small bubbles) is better at controlling fouling in submerged hollow fiber membrane systems, when compared to sparging with coarse bubble diffusers for a given flow rate [132, 176]. Considering that the size of the diffuser nozzle used in the present study did not significantly impact the magnitude of the average surface shear signal, the results suggest that the better fouling control associated with sparging with smaller bubbles is likely related to the number of surface shear events that the fibers experience. The higher number of surface shear events generated by small bubbles (i.e. small diffuser nozzle) in low and medium fiber packing density bundles likely provides more favorable hydrodynamic conditions for fouling reduction and flux enhancement.

5.3.1.2 Shear Profile During Surface Shear Event

As presented in Figure 5-6, the geometry of the bubbles rising between fibers was substantially affected by the experimental conditions (i.e. diffuser nozzle size, gas sparging rate and fiber packing density).

Contribution of Rising Bubbles to Positive Peak Surface Shear Signal

For the experiments in which the surface shear signals were consistently positive (e.g. graph aM, Figure 5-3), an analysis of the images shows that the positive shear peaks were caused by the wake generated at the tail end of a bubble as the bubble rose past the shear probe. Figure 5-8 shows a typical shear profile of a tightly held fiber bundle during the passage of a bubble, and the corresponding bubble images. As the bubble approached the location of the shear probe on the test-fiber, the shear signal began to rise rapidly (points 1 and 2, Figure 5-8)⁴. When the tail end of the bubble passed by the shear probe, a positive peak in the surface shear signal occurred (point 3, Figure 5-8). The decrease in surface shear signal after this point occurred relatively slowly, even though the bubble was no longer in the vicinity of the shear probe (point 4, Figure 5-8). These results suggest that the wake region of the rising bubble was long, and that the secondary flows in the wake region likely play an important role in contributing to particle back-transport at a membrane surface. This observation is consistent with results from other studies that indicate that the wake structure of a bubble rising in water at a relatively low Reynolds number is larger than the size of the bubble itself [178].

 $^{^4}$ It has been reported that the electrochemical shear probe could measure fluctuations in flow of up to 100 Hz [177], which corresponds to 0.01 seconds. The peak observed in Figure 5-8, i.e. between points 1 and 3) occurred over a period of 0.03 seconds. As such, the probe was likely capable of detecting the peak in Figure 5-8.



Figure 5-8. Typical shear profile and images of a spherical bubble rising in tightly held fibers

The translucent vertical streaks seen in the pictures are the 8 fibers in the bundle

The black dashed line highlights the bubbles rising within the fiber bundle. The white horizontal dashed line across the 4 images shows the position of the shear probe on the test-fiber. (Fiber packing density: medium; gas sparging rate: 0.5 mL/min, diffuser nozzle size 2 mm)

When examining the surface shear signals in Figure 5-3 and Figure 5-4, the majority of the surface shear events were positive and similar to the one seen in Figure 5-8. The results presented in Figure 5-8were for a spherical bubble rising parallel to the test-fiber. However, depending on the system configuration and the gas sparging rate, other bubble geometries were

also observed, as presented in Figure 5-6. The geometry of the bubbles had a significant impact on the surface shear signals. Ellipsoidal bubbles were commonly observed when the gas sparging rate was high. Figure 5-9 shows a typical shear profile resulting from the passage of an ellipsoidal bubble by the shear probe as the bubble 'escaped' from a tightly held fiber bundle. Again, the maximum surface shear signals were observed at the tail end of the bubble (i.e. after the bubble had passed by the shear probe). However, in contrast to the peak surface shear signal presented in Figure 5-8, which lasted only 1 to 2 milliseconds, the peak in the surface shear signal observed in Figure 5-9 lasted approximately 20 milliseconds (points 4, 5 and 6 in Figure 5-9). The different shear profiles observed for the different bubble geometries likely have different effects on fouling control. Whether the high frequency of peaks similar to that seen in Figure 5-8, or the longer duration of high surface shear signal seen in Figure 5-9 are the most favorable for flux enhancement, is unknown. The results from a study that investigates the relationship between different types of transient shear profiles and fouling is presented in Chapter 8.

Contribution of Rising Bubbles to Negative Peak Surface Shear Signals

One of the mechanisms of fouling reduction for slug flow inside a confined membrane system has been linked to the high negative shear stresses induced by the liquid flow reversal (falling film) surrounding a rising gas slug [10]. In order for flow reversal to occur, the flow path of the gas slug must be confined. As presented in Figure 5-3 and Figure 5-4, negative surface shear signals were periodically observed, suggesting that liquid flow reversal periodically occurred during the passage of bubbles by the shear probe, especially for the experiments using a 2 mm diameter diffuser nozzle, a high gas sparging rate and a high fiber packing density. The number of negative surface shear events, compared to the total number of surface shear events measured for each experiment, is tabulated in Table 5-2.



Figure 5-9. Typical shear profile and images of an ellipsoidal bubble rising in tightly held fibers

The translucent vertical streaks seen in the pictures are the 8 fibers in the bundle. The white horizontal dashed line across the 6 images shows the position of the shear probe on the test-fiber. (Fiber packing density: high; gas sparging rate: 0.5 mL/min, diffuser nozzle size 2 mm)

Experiment No.	Packing Density	Nozzle Size (mm)	Gas Flow (mL/min)	Total Number of Surface shear events	Number of Negative Peaks	% of Negative Peaks out of Total Number of Surface shear events
1-A1	Low	1	2	11	1	9
1 - B1	Med	1	2	10	0	0
1-C1	High	1	2	8	8	100
2-A1	Low	2	2	10	3	30
2-B1	Med	2	2	9	0	0
2-C1	High	2	2	9	0	0
1-A2	Low	1	10	56	4	7
1-B2	Med	1	10	68	1	1
1-C2	High	1	10	24	1	4
2-A2	Low	2	10	36	1	3
2-B2	Med	2	10	49	0	0
2-C2	High	2	10	47	1	2
1-A3	Low	1	35	85	0	0
1-B3	Med	1	35	208	2	1
1-C3	High	1	35	101	2	2
2-A3	Low	2	35	43	0	0
2-B3	Med	2	35	171	21	12
2-C3	High	2	35	146	45	31

Table 5-2. The number of negative peaks observed in Figure 5-3 and Figure 5-4

It can be seen from Table 5-2 that, with the exception of experiment 1-C1, the majority of the surface shear signals were positive in value, indicating that a reversal in the direction of liquid flow typically did not occur frequently at the surface of the test-fiber. It is noted here that this observation is different than that of the shear profiles of rising gas slugs in confined systems such as tubular membranes, where each surface shear event consists of both positive and negative shear stresses. For the positive peaks, the rising bubbles were relatively close to the surface of the test-fiber. However, because of the packing density and the flexibility of the fibers, as well as the different bubble sizes, the sparged bubbles were typically not completely confined to rise against the surface of the test-fiber, as would be the case for a gas slug rising in a confined system. As a result, flow reversal did not consistently occur at the surface of the test-fiber. However, at higher fiber packing densities or at higher gas sparging rates, periodic flow reversal was observed. These results are consistent with the images in Figure 5-6b and Figure 5-6c, where the bubbles rising between fibers were periodically observed to be similar in shape to that of a gas slug rising in a confined tubular membrane system.

As presented in Figure 5-10, for the negative surface shear events, the shear profile was different from those presented in Figure 5-8 and Figure 5-9. Although the bubbles presented in Figure 5-8 and Figure 5-9 were largely spherical, the bubble shown in Figure 5-10 was elongated like a gas slug. The difference in bubble shapes was due to the differences in the geometry of the flow path of the rising bubble. The bubble seen in Figure 5-10 was relatively confined within a high fiber packing density bundle. Therefore, the rising bubble assumed a shape similar to that of a gas slug rising in a confined vertical tube. On the other hand, the bubbles presented in Figure 5-8 and Figure 5-9 were spherical because they were not confined by a high fiber packing density. Two positive and two negative peaks in surface shear signals were observed in Figure 5-10. The first positive peak (point 2, Figure 5-10) occurred as the nose of the bubble passed by the shear probe. After the first positive peak in surface shear signals, two negative peaks were observed at approximately 10 and 38 milliseconds after the passage of the tip of the bubble past the shear probe. The images taken at these negative peaks in surface shear signals showed that the shear probe was in the body of the rising bubble, suggesting that the negative surface shear signals were due to liquid flow reversal in this region. The second positive peak in surface shear signals were due to liquid flow reversal in this region.

signals (point 5, Figure 5-10) occurred near the tail end of the bubble as it rose past the shear probe. The positive surface shear signals that were observed between points 3 and 4 (Figure 5-10) likely resulted from the incompletely confined nature of the flow path of the rising bubble and therefore a non-stable falling film. Negative surface shear signals were also periodically observed when the tail end of rising bubbles passed by the shear probe. These results suggest that some of the negative surface shear signals were generated by the secondary flows induced by the wake of a rising bubble. The negative surface shear signals generated at the tail end of rising bubbles are shown in Figure 5-11. As presented, positive peak surface shear signals (point 2 on Figure 5-11) were immediately followed by a negative peak surface shear signal. Both the positive and negative peak surface shear signals occurred when the tail end of the bubble passed by the shear probe, suggesting that the oscillating surface shear signals resulted from the secondary flows generated in the wake region of a rising bubble⁵. It should be noted that the bubble images presented in Figure 5-11 are similar to those presented in Figure 5-8, but the shear profiles for the two bubbles were very different. The video imaging system used was only capable of capturing a two dimensional view of the bubble relative to the location of the shear probe. It is likely that the differences in the shear profiles presented in Figure 5-8 and Figure 5-11 were due to the differences in the distance between the bubbles and the shear probe in the third dimension, which could not be imaged.

A closer examination of the shear profiles for the experiments in which negative surface shear signals were observed revealed that, on average, the frequency at which negative surface shear signals were observed either at the nose or tail regions of a rising bubble passed by the shear probe, were similar. These results indicate that for the experimental conditions investigated, negative surface shear events were equally likely to occur due to flow reversal or due to the secondary flows in the wake of a rising bubble.

 $^{^{5}}$ The single negative peak (point 3) in Figure 5-11 occurred over 0.001second. It is noted here that it may be possible that the shear probe was not able to detect such a high frequency fluctuation, and that the negative peak is a product of instrument artifact as opposed to being real. The negative peaks in Figure 5-10 lasted longer – those peaks are likely real.



Figure 5-10. Typical shear profile and images of a bubble causing negative shear peaks due to falling film surrounding the bubble

The translucent vertical streaks seen in the pictures are the 8 fibers in the bundle. The black dashed line highlights the bubbles rising within the fiber bundle. The white horizontal dashed line across the 5 images shows the position of the double probe on the test-fiber. (Fiber packing density: high, gas sparging rate: 2 mL/min, diffuser nozzle size: 1 mm, configuration: tightly held)



Figure 5-11. Typical shear profile and images of a sparged bubble causing negative shear peaks due to flow reversal generated by instabilities in the wake region

The translucent vertical streaks seen in the pictures are the 8 fibers in the bundle. The black dashed line highlights the bubbles rising within the fiber bundle. The white horizontal dashed line across the 4 images shows the position of the shear probe on the test-fiber. (Fiber packing density: medium, gas sparging rate: 35 mL/min, diffuser nozzle size: 2 mm, configuration: tightly held)

5.3.2 Bubble-Induced Surface Shear Profile for Loosely Held Fiber Bundles

Experiments with loosely held fibers at a gas sparging rate of 35 mL/min were conducted for high fiber packing density bundles, and for diffuser nozzle sizes of 1 mm and 2 mm. Typical surface shear signals for loosely held fibers are shown in Figure 5-12. All of the surface shear signals observed in Figure 5-12 were positive, indicating that for loosely held fibers, gas sparging did not result in any flow reversal. This implies that scouring of the fiber surface by a falling film, which is an important mechanism in the control of fouling in confined membrane systems [10], does not occur in loosely held submerged hollow fiber systems.



Figure 5-12. Typical surface shear signals on loosely held fibers

Fiber packing density: high, gas sparging rate: 35 mL/min, diffuser nozzle size 2 mm. The zoomed-in view of the circled section, along with images of the rising bubble, are shown in Figure 5-13

As presented in Figure 5-12, the average surface shear signals were, in general, lower than those observed for the tightly held fibers. However, occasionally, larger peaks in surface shear signal were observed. Video images shown in Figure 5-13, revealed that most of the time, bubbles were not in contact with the test-fibers, since the flow path was not as confined as was the case for tightly held fibers. This resulted in the overall lower surface shear signals. Occasionally, bubbles came very close to the shear probe, causing larger peaks in surface shear signals. However, the frequency at which these larger peaks in surface shear signals occurred was relatively low compared to the frequency at which bubbles were released from the diffuser. Despite this, a number of studies have indicated that higher permeate flux can be maintained in a loosely held submerged hollow fiber membrane system, compared to a system with tightly held fibers [86, 140]. This discrepancy can likely be explained by the fact that bubbles rising in systems with tightly held fibers are relatively confined to a specific region of the fiber bundle. In contrast, for systems with loosely held fibers, the flow paths of the bubbles are not confined to a specific region. Also, as the bubbles rise past the loosely held fibers, secondary flows generated by the wake of the rising bubbles can cause the fibers to sway [107, 140]. The swaying of the loosely held fibers in turn increases the likelihood that bubbles will travel to multiple regions of the loosely held fiber bundle, thus increasing the number of fibers that can benefit from the bubble-induced shear stresses. The results from studies that investigated the distribution of shear within a bundle are presented in Chapter 7.



Figure 5-13. The zoomed-in view of the circled section in Figure 5-12, along with the synchronized images of the rising bubble

The translucent vertical streaks seen in the pictures are the 8 fibers in the bundle. One of the bubbles in Frames 2221, 2228, 2701 and 2713 is partially behind the translucent fiber. Fiber packing density: high; gas sparging rate: 35 mL/min, diffuser nozzle size 2 mm, configuration: loosely held

5.4 Conclusions

Shear profiles during sparging were found to be substantially affected by the operating conditions (i.e. gas sparging rates, fiber packing density, diffuser nozzle size, and module configuration). Based on the experimental data, the following conclusions were reached.

- The hydrodynamic conditions in confined tubular membrane systems and unconfined submerged hollow fiber membranes were observed to be different. These results imply that, in contrast to the situation with confined tubular membranes, flux enhancement in gas-sparged submerged hollow fiber membrane systems is likely not achieved through scouring of the fiber surface by a falling film.
- Different operating conditions yielded bubbles with different geometries (i.e. spherical, ellipsoidal or slug-like), and the different bubble geometries yielded different shear profiles.
- 3. Scouring of the fiber surface by a falling film, which is an important mechanism in the control of fouling in confined membrane systems, may not occur in loosely held submerged hollow fibers.
- 4. The flow path of bubbles rising in loosely held fibers was not confined to a specific region, as was the case for tightly held fibers. The larger number of fibers (i.e. regions) that can benefit from sparged bubbles in systems with loosely held fibers likely explains why higher permeate fluxes have been reported in loosely-held, submerged, hollow-fiber membrane systems, compared to those in tightly held systems.
6. Shear Stress at Radial Sections of a Submerged Hollow Fiber Under Gas Sparging

6.1 Introduction

The results from the experiments presented in this chapter are an extension of those described in Chapter 5, in which surface shear signals of the fiber section facing the direction of the sparged gas bubble was investigated. It is not known if the different radial sections of the fiber, not directly facing the sparged bubble, experience the same magnitude of surface shear signal as the sections that directly face the sparged bubble. In this chapter, the results from a study that investigated the directional shear stress acting on different radial sections of a tightly-held fiber resulting from two-phase (gas-liquid) flow, are presented for different gas flow rates, and for gas delivered through gas nozzles of different sizes. Surface shear signal measurement was based on the electrochemical method described in Chapter 5.

6.2 Materials and Methods

In this section, only the summary of the experimental set-up is presented. A detailed description of the experimental set-up used is given in Section 5.2. Shear probes mounted on the test-fiber (as described in Chapter 4 and 5 were used.) The bench-scale hollow fiber unit consisted of a bundle of 8 fibers arranged in a 3 x 3 square matrix. The fibers were tightly held. The test-fiber, on which the shear probe was mounted is presented in Figure 5-2b. Gas was delivered through the gas nozzle positioned at the center of the bundle. The position of the shear probe relative to the rising bubble at the center of the bundle was adjusted by rotating the test-fiber clock-wise and counter-clock-wise, as presented in Figure 6-1. A zero degree rotation indicated that the shear probe was directly facing the rising bubble, while a 180 degree rotation indicated that the shear probe was facing away from the rising bubbles. Such an arrangement allowed for the measurement of the surface shear stress at different radial sections on the test-fiber. Shear profile measurements were conducted for gas delivered through gas diffusers with nozzle sizes

of 1 mm and 2 mm, and gas flow rates of 4 mL/min, 30 mL/min and 35 mL/min. Data was measured/acquired at rate of 1000 Hz, for 10 seconds for each experiment. The measured signal was corrected for instrument signal bias, as described in Section 4.3.6. All experiments were performed in replicates.



(a) Side View

(b) Top View

Figure 6-1. Bundle of 8 fibers

(a) Side view. (b) Top view of probe orientation, relative to the rising gas bubble. θ angle of rotation. $\theta = 0^{\circ}$ – shear probe facing the bubble. $\theta = 180^{\circ}$ – shear probe facing away from the bubble.

6.3 Results

6.3.1 Typical Shear Profiles at Radial Sections of the Fiber Surface

Typical shear profiles of different radial sections of the tightly-held fiber (from $\theta = 0^{\circ}$ to 180°) during gas sparging through a gas nozzle size of 1 mm, and at a gas flow rate of 35 mL/min are shown in Figure 6-2. Similar to the shear profiles observed in Figure 5-3, shear profiles for radial sections from 0° to 135° were characterized by a number of positive peaks in surface shear signal. Note that the magnitude of the surface shear signals in Figure 6-2 were lower than those observed in Figure 5-3cM (for the same fiber packing density, gas flow rate and gas nozzle size). This was likely due to the fact that a different shear probe was used for the experiments conducted in Figure 6-2. As discussed in Section 4.3.5, the surface area of each probe is slightly different, resulting in different signal magnitudes unless calibrated or normalized. However, since the analysis focuses on trends in surface shear signals, rather than the absolute surface shear signal, the results presented still provides insight on the effect of radial sections of the surface shear signals.

As discussed in Section 5.3, each of the peak, or surface shear event likely resulted from the high shear stress induced by the wake of a sparged bubble. Numerous positive surface shear events were also observed in the shear profiles for experiments conducted at lower gas flow rates of 5 mL/min, 35 mL/min, and a gas nozzle size of 2 mm (see Appendix H). The shear profiles (i.e. number of peaks, average magnitude of peaks, maximum peak value, average surface shear signal), however, varied for experiments with different gas flow rates and gas nozzle sizes. Since particle back-transport from membrane surfaces is dependent on shear stresses experienced at the wall of the membranes [10, 179], the shear profile is expected to play an important role in the control and prevention of fouling on membrane surfaces. The variation in the shear profiles for different gas flow rates and gas nozzle size for different gas flow rates and gas nozzle size save discussed in the following sections.











(e)

Figure 6-2. Typical bubble-induced shear profiles at different radial sections of the fiber surfaces

a) $\theta = 0$ degrees, b) $\theta = 45$ degrees, c) $\theta = 90$ degrees, d) $\theta = 135$ degrees, e) $\theta = 180$ degrees Data shown for a gas flow rate of 35 mL/min, delivered through gas nozzle size of 1 mm

6.3.1.1 Comparison of Shear Profiles for Different Gas Flow Rates

The number of shear events in the shear profile at different radial sections, and for different experiments are shown in Figure 6-3a and Figure 6-3b. The number of shear events, experienced by the fiber at different radial sections during gas sparging was found to be similar for all radial sections of the fiber, with the exception of the sections at 180° and -180°. In this section, no desirable shear event was observed in the shear profile. This suggests that the surfaces of fiber radial sections of 135° and 180° were largely shielded and did not experience oscillatory and high shear flows, compared to the other sections. Also, as expected, the numbers of shear events were higher for experiments conducted at higher gas flow rates (30 and 35 mL/min, compared to 4 mL/min). This is consistent with the results reported in section 5.3 where higher gas flow rates result in the formation of greater numbers of shear events.



Figure 6-3. Comparison of the number of shear events per 10 seconds of data in the shear profile

(a) 2 mm gas diffuser, (b) 1 mm gas diffuser. Error bars represent confidence intervals equivalent to one standard deviation of the replicate experiments. Note the standard deviation of the data points in Figure 6-3b is small, the error bar is not visible on the graph

The average magnitude of the peaks $(\overline{\tau_{peak}})$ was calculated (Figure 6-4), the maximum magnitude of all peaks (max_{peak}) , as well as the average magnitude of all surface shear signals $(\overline{\tau_{all}})$ for the different experiments are shown in Figure 6-4, Figure 6-5 and Figure 6-6

respectively. Although the differences between these values at the radial sections considered are not statistically significant, the following trends were nonetheless consistently observed:

- the highest $\overline{\tau_{peak}}$, max_{peak} and $\overline{\tau_{all}}$ were found in the fiber sections closest to rising bubbles, and these decreased substantially at radial sections further away from the rising bubbles,
- at the radial sections of 180° on the test-fiber, $\overline{\tau_{peak}}$ and $\overline{\tau_{all}}$ were zero, and
- $\overline{\tau_{peak}}$, max_{peak} and $\overline{\tau_{all}}$ were consistently greater at higher gas flow rates.

The surface shear signals between 0° and -180° , and 0° and 180° were not symmetrical (i.e. skewed as shown in Figure 6-4). The reason for this asymmetry is likely due to the gas sparger used, which resulted in a skewed flow path of the bubbles (i.e. bubbles tended to rise towards radial sections 0° and -180° of the fiber.) Since the frequency of occurrence of the high shear conditions mostly experienced at radial sections from 0° to 135° , the permeate flux enhancement as a result of bubble-induced shear would be expected to be most significant in these sections of the fiber. Radial sections 135° to 180° may experience the least permeate flux enhancement by bubble-induced shear.



Figure 6-4. The average value of all peaks ($\overline{\tau_{peak}}$) in the shear profiles of each experiment

(a) 2 mm gas diffuser, (b) 1 mm gas diffuser. Error bars represent confidence intervals equivalent to one standard deviation of the replicate experiments



Figure 6-5. The maximum peak value of all shear events (max_{peak}) in the shear profiles

(a) 2 mm gas diffuser, (b) 1 mm gas diffuser. Error bars represent +/- standard deviation of the replicate experiments



Figure 6-6. Average surface shear signals ($\overline{\tau_{all}}$) of all shear profiles

(a) 2 mm gas diffuser, (b) 1 mm gas diffuser. Error bars represent +/- standard deviation of the replicate experiments.

6.3.1.2 Comparison of Shear Profile for Different Gas Nozzle Sizes

Figure 6-7 and Figure 6-8 show the comparison between experiments with sparging using the 1 mm and 2 mm gas nozzles in terms of $\overline{\tau_{peak}}$, max_{peak} and $\overline{\tau_{all}}$ at radial sections from 0° to 180°. Although the differences between these values at the radial sections considered are not statistically significant, however, the number of peaks counted in the shear profile was nonetheless found to be consistently higher in the experiment sparged with the 1 mm gas nozzle, compared to the 2 mm gas nozzle (Figure 6-7a and Figure 6-8 a) for fiber radial sections from 0° to 135°. This observation is consistent with that reported in Chapter 5, and is also consistent with published literature [179].

In terms of maximum peak value, as well as the calculated average surface shear signal, the differences in the values for the 1 mm and 2 mm gas nozzle were found not to be statistically significant, for all radial sections. However, this observation does not preclude the fact that the calculated average signals for the 1 mm and 2 mm experiments were similar, rather it is possible that the experimental set-up used in this study is not sensitive enough to capture the differences. As discussed in Section 3.6, sparging with larger bubbles can potentially generate shear events of greater magnitude [104, 131]. It is possible that the selection of the optimum nozzle size may depend on the competing benefits of increased numbers of peaks experienced by the majority of radial sections of the fiber induced by smaller gas bubbles (i.e. smaller gas nozzle), and the greater shear experienced by a smaller section of fiber induced by larger gas bubbles (i.e. *larger* gas nozzle). The latter case, although beneficial to only a small section of the fiber, can result in non-uniform membrane surface fouling. Depending on the application of the hollow fiber membrane, and the water/wastewater matrix filtered, non-uniformity in fouling at different sections of the fibers has been linked to decreased permeability of the fiber bundle as a whole due to inter-fiber caking [109]. As such, it is possible that the increased number of peaks generated by smaller bubbles sparged through a smaller nozzle is more beneficial in terms of uniform fouling control and overall flux enhancement, when compared to sparging using larger gas nozzles. This hypothesis is consistent with the observations of Sofia [132] and Fane [176].



Figure 6-7. Number of shear events at different radial sections of a hollow fiber, at a gas flow rate of 4 mL/min

a) Number of peaks, b) peak average, c) maximum peak value, d) average surface shear signal. Error bars represent confidence interval equivalent to one standard deviation of the replicate experiments.



Figure 6-8. Number of shear events at different radial sections of a hollow fiber, at a gas flow rate of 35 mL/min

a) Number of peaks, b) peak average, c) maximum peak value, d) average surface shear signal. Error bars represent confidence interval equivalent to one standard deviation of the replicate experiments.

6.4 Conclusions

The directional surface shear signal acting on different radial sections of a fiber resulting from two-phase (gas-liquid) flow, was measured for different gas flow rates, and for gas delivered through gas nozzles of different sizes. It was found that the sections of the fiber closest to the sparged bubble experienced surface shear events of the highest magnitude, although the total number of surface shear events experienced by different radial sections of the fiber was similar. These results indicate that only a small portion of the fibers benefit from the surface shear events induced by rising sparged bubbles. This suggests that significant shielding possible occurs, where fibers not immediately adjacent to the gas bubbles do not experience the same magnitude

of surface shear signals, compared to fibers located immediate adjacent to the gas bubbles. Chapter 7 is dedicated to investigating the shielding effect in a bundle of fibers, when sparging is located outside of the fiber bundle.

7. Distribution of Shear Inside a Gas-Sparged Hollow Fiber Bundle

7.1 Introduction

The effective distribution of the bubble-induced shear stresses across membrane surfaces is important for optimized use of gas sparging for fouling control. The distribution of these shear stresses inside the membrane module is influenced by the location of the gas nozzle, the distribution of fibers within the membrane module, and the fiber packing density. Fibers that are located in the outer sections of a module of hollow fibers may experience the benefits of the high shear stresses generated by a rising sparged gas bubble to a greater extent than fibers that are located within the module, especially if the packing density is high. Depending on the packing density, the hydrodynamic conditions inside the bundle can be very different than the hydrodynamic conditions outside the bundle [108, 109]. Results from Chapter 6 also suggest that only the areas of the fibers directly facing the bubble, or those that faced the bubbles will benefit from the bubble-induced shear stresses. Additionally, in systems where gas sparging occurs either intermittently or continuously, movement of hollow fibers has been observed [140, 180]. Long fibers move more frequently, and at a higher amplitude, than shorter fibers [142]. Improvement in fouling control has been attributed to fiber movement in loosely-held fibers [44, 140, 141]. The fiber movement may promote physical contact between membrane surfaces, which may result in scouring of membrane surfaces. The approach used for gas sparging also plays an important role in improving fouling control. Several authors have reported that there is a critical gas sparging flow rate above which increasing the sparging flow does not result in a further improvement in fouling control in submerged membrane systems [44, 71, 104, 127, 181].

The aim of the study described in this chapter was to provide a more detailed investigation of the hydrodynamic conditions that exist inside a gas sparged bundle of hollow fiber membranes, when gas bubbles were delivered outside of the bundle. Specifically, the effects of shielding of fibers within a bundle were examined. This was achieved by mapping the surface shear signal distribution on the surfaces of a bench-scale hollow fiber bundle under typical gas sparging

conditions. The effect of gas nozzle size, nozzle location, gas sparging rate, fiber configuration, as well as fiber movement on the distribution of the surface shear signal within the bundle were also examined.

7.2 Materials and Methods

The distribution of shear stresses acting on the surfaces of fibers at different locations within a bench-scale hollow fiber membrane bundle was investigated. Shear stresses acting on membrane surfaces were measured using an electrochemical method. Different operating conditions and membrane configurations, such as gas diffuser size, gas diffuser location (i.e. distance from fibers), gas flow rate, and fiber tension were studied. The effects of fiber movement and physical contact between fibers were also investigated.

7.2.1 Electrochemical Shear Measuring System

The electrochemical shear measuring system was similar to that described in Section 5.2. A shear probe was embedded flush to the surface of the test-fibers. The test-fibers were made of Teflon tubes (1.7 mm o.d) with similar dimensions and flexibility as those of a hollow fiber membrane. However, unlike the shear probes described in Section 5.2, which consisted of two probes placed side by side, only one probe was mounted on the test-fiber in this study. This single-probe system was chosen for two reasons:

- 1. It is easier to mount a single probe on the test-fiber compared to mounting two probes on the test-fiber,
- Results described in Chapter 5 suggested that the occurrence of flow reversal near membrane surfaces was limited. Therefore, the single-probe, which is only capable of measuring the magnitude of shear, and not the direction of shear, was considered sufficient for the experiments conducted in this study.

The procedure for the mounting of a single probe onto the test-fiber was different than that for the two-probe. A 0.5 mm hole was first drilled on the surface of the test-fiber. A piece of

platinum wire was inserted through the hole from the underside of the test- fiber. Epoxy was injected into the cavity of the test-fiber to secure the platinum wire in place. The protruding platinum wire was then sanded down such that its surface was flush with that of the test-fiber. More details of the mounting procedure can be found in [182]. The shear probes were placed 3 cm from the bottom end of the vertically oriented test-fiber.

As discussed in Section 4.3.1, due to the nature of the manufacturing process, the surface area of each probe was slightly different. As a result, for a given hydrodynamic condition, the surface shear signals measured by each probe differed slightly. To ensure that the signals from each probe could be compared, all the probes were normalized to the surface shear signal measured from the probe on one test-fiber. Details of the normalization procedures are provided in Appendix I.

The electrolytic solution for the shear measuring system (which contains potassium ferricyanide, potassium ferrocyanide and potassium chloride), and the data acquisition system were described in detail in Sections 4.3.3 and 4.3.1 respectively .

7.2.2 The Bench-Scale Fiber Bundle, System Tank, and Gas Sparging System

Nine test-fibers were arranged in a 3 x 3 square matrix, as shown in Figure 7-1. Each test-fiber was 12 cm in length, and was identified as P1, P2, P3, P4, P5, P6, P7 and P_{no} . Shear probes were embedded flush on the surface of P1 to P7. P_{no} did not have shear probes embedded (see Figure 7-1). It was initially assumed that due to the geometric symmetry of the bundle, the hydrodynamic conditions near P_{no} would be similar to those near P2 and P4 when nozzles A and B were used. The probes on each test-fiber faced the y-direction, as indicated on Figure 7-1.



Figure 7-1. Arrangement of the test-fibers in a 3 x 3 square matrix

Test-fibers with shear probes embedded were identified as P1 to P7. The gas nozzles A, B, D and E were located adjacent to the bundle of test-fibers

The spacing between the test-fibers was 1 mm. Immediately adjacent to the matrix of the test-fibers were four different gas nozzles, (A, B, D, and E). Fibers P1 and P2 were closest to the gas nozzles, while P5 to P7 were farthest away from the gas nozzles. The spacing between the gas nozzles, and the spacing between the fibers closest to nozzles A and D (i.e. P1 and P2) were both 1 mm. The centerline of nozzles A and D coincide with the centerline of test-fibers P1, P3 and P6

Similar to the experiments described in Section 5.2, the fiber bundles were immersed in a cylindrical system tank filled with the electrolyte solution. Both ends of the bundle (i.e. circular discs) were held in place such that the bundles could be held either loosely or tightly by adjusting the height at the top of the bundle. For tightly held bundles, the distance between the circular disks which held the ends of the fibers was 12 cm. For loosely held fibers, the distance between the circular the circular disks was 95 % of this value (i.e. 11.4 cm).

The cylindrical system tank was constructed of plexiglas so that bubbles in the tank could be observed visually. The system tank was sealed at the top to prevent oxygen exposure during each experiment. The nickel anode was also immersed in the system tank.

Nitrogen gas was delivered adjacent to the test-fibers through gas nozzle sizes of 1 mm or 3 mm (i.d.). Nitrogen gas, instead of air, was used because ferricyanide and ferrocyanide reacts chemically with oxygen.

7.2.3 Simulation of Fiber Movement and Physical Contact Between Fibers

The effect of fiber movement on shear distribution within the bundle was studied. Due to the relatively short length of the test-fibers in the bundle, fiber movement had to be simulated manually. Fiber movement was simulated by physically moving the test-fiber bundle in pendulum motion in the system tank, as shown in Figure 7-2. One complete movement from left to right was considered one cycle. The test-fiber bundle was fixed at the bottom, while the top section was free to move 2 cm or 4 cm from the center of the axis. The frequency of the cycles was approximately 0.5 Hz for both the 2 cm and 4 cm amplitudes. Therefore, test-fibers moved at a faster velocity for the experiment with the 4 cm amplitude, compared to those with the 2 cm amplitude. For the investigation of the effect of physical contact between fibers, the test-fibers were held loosely. Contact between fibers was simulated by manually "touching" the shear probe on the test-fiber surface to another test-fiber.



Figure 7-2. Simulation of fiber movement

7.2.4 Experimental Program

An experimental program which considered different module configurations (loosely and tightly held), diffuser locations (A, B, D and E), diffuser size (1mm and 3 mm), and gas sparging rates (10, 40, 55, 95, 142, 195 mL/min) was conducted to quantify the distribution of shear stresses inside a fiber bundle. A summary of all experiments performed is presented in Table 7-1 All experiments were performed in random order. In addition to the experiments listed in Table 7-1, the experiments that investigated the effects of fiber movement and contact between fibers were also performed.

Nozzle Size	Nozzle	Gas Flow	Fiber	Nozzle Size	Nozzle	Gas Flow	Fiber
(mm)	Location	(mL/min)	Configuration	(mm)	Location	(mL/min)	Configuration
1	А	10	Tightly held	3	А	10	Tightly held
1	А	40	Tightly held	3	А	40	Tightly held
1	А	55	Tightly held	3	А	55	Tightly held
1	А	95	Tightly held	3	А	95	Tightly held
1	А	142	Tightly held	3	А	142	Tightly held
1	А	195	Tightly held	3	А	195	Tightly held
1	А	195	Loosely held	3	А	195	Loosely held
1	В	10	Tightly held	3	В	10	Tightly held
1	В	40	Tightly held	3	В	40	Tightly held
1	В	55	Tightly held	3	В	55	Tightly held
1	В	95	Tightly held	3	В	95	Tightly held
1	В	142	Tightly held	3	В	142	Tightly held
1	В	195	Tightly held	3	В	195	Tightly held
1	D	10	Tightly held	3	D	10	Tightly held
1	D	40	Tightly held	3	D	40	Tightly held
1	D	55	Tightly held	3	D	55	Tightly held
1	D	95	Tightly held	3	D	95	Tightly held
1	D	142	Tightly held	3	D	142	Tightly held
1	D	195	Tightly held	3	D	195	Tightly held
1	Е	10	Tightly held	3	Е	10	Tightly held
1	Е	40	Tightly held	3	Е	40	Tightly held
1	Е	55	Tightly held	3	Е	55	Tightly held
1	Е	95	Tightly held	3	Е	95	Tightly held
1	Е	142	Tightly held	3	Е	142	Tightly held
1	Е	195	Tightly held	3	Е	195	Tightly held

Table 7-1. Summary of the experimental program

All experiments were conducted at room temperature (23 °C) and under limiting current condition. The applied potential between the anode and cathode was 300 mV. Ten seconds of data were recorded during each experiment at an acquisition rate of 1000 Hz. The recorded signals were corrected for instrument bias and normalized. The time averaged surface shear signal, the standard deviation of the surface shear signal, and the total number of shear events (surface shear signal peak count) from each experiment are reported. The appropriateness of the parameters such as time averaged shear and standard deviation in describing the hydrodynamic conditions in a gas-sparged system, and their relationship to fouling control is discussed in detail in Chapter 8. For the present study, however, the time averaged surface shear signal and the

standard deviation of shear signal was used to compare the results from the different experimental conditions investigated.

All experiments were performed in duplicate. Due to the limitation of the experimental apparatus, it was often difficult to control the path of the bubbles rising. Therefore, the results were not identical for the different replicate experiments. For this reason, confidence intervals in the form of error bars are not presented on the figures in the following sections. However, certain trends observed in the experiments were repeatable for the duplicate experiments. Emphasis is placed on the repeatable trends observed.

As discussed in Section 5.2, surface shear signals (V) are presented instead of calculated shear stress values (Pa) to avoid introducing any bias into the experimental results, as well as propagating errors.

7.3 Results and Discussions

7.3.1 Tightly-held Bundle

7.3.1.1 Typical Shear Profiles

Typical shear profiles measured on test-fibers P1 to P7, when sparging through gas nozzle A, are presented in Figure 7-3. The shear profiles were characterized by spikes in surface shear signals over time (i.e. shear events). For test-fibers P1 and P2, which were located closest to the gas nozzle, the magnitude and variability (in terms of standard deviation) of the surface shear signals were higher than for other test-fibers (surface shear signals between 0.4 and 1 V). During gas sparging, the distance between the probes on test-fibers P1 and P2 and the rising bubbles were observed to be approximately 1 to 2 cm. The high surface shear signals (greater than 0.4 V) were likely the result of the wake generated at the tail end of the bubble rising past the shear probe on test-fibers P1 and P2 (see discussion in Section 5.3.1.2). Occasional surface shear signals higher than 0.9 V were also observed (Figure 7-3). These higher peaks are likely due to bubble passing in relatively closer proximity to the shear probes on test-fibers P1 and P2. For test-fibers P3 to P7, which were located behind test-fibers P1 and P2, the magnitude and

variability of the surface shear signals was lower than for test-fibers P1 and P2. The surface shear signals of test-fibers P6 to P7, which were the furthest from the nozzles, were even lower than that for test-fibers P4 and P5. These results suggest that fibers not immediately adjacent to the gas nozzle were not significantly impacted by the sparging bubbles (i.e. shielded). This is consistent with results presented in Chapter 6 which indicated that only a small portion of the fibers benefit from the surface shear events induced by rising sparged bubbles closest to the fiber.



Figure 7-3. Typical shear profiles measured by probes P1 to P7, when sparging though gas diffuser A

Gas flow rate: 195 mL/min; gas nozzle size: 3 mm; bundle configuration: tightly-held

The magnitude and variability of surface shear signals measured on test-fibers P1 and P2, when sparging through gas nozzles B, D and E, were substantially lower than those measured when sparging through nozzle A. This difference is illustrated Figure 7-3 and Figure 7-4. For test-fibers P1 and P2, which were located closest to the gas nozzle, the magnitude and variability of the surface shear signals were again higher (surface shear signals between 0.4 and 1V) than those

of test-fibers P3 to P7. However, when compared to the shear profiles presented in Figure 7-3, the magnitude and the variability of the surface shear signals were substantially lower. This suggests that the width of the wakes generated at the tail end of the bubbles rising further away from the fibers were not wide enough to induce substantial surface shear signals on the test-fibers. Occasional higher peaks in surface shear signals were observed for test-fibers P1 and P2 (between 0.4 and 0.6 V). These peaks were likely the result of bubbles periodically passing in closer proximity to the shear probes on the test-fibers.



Figure 7-4. Typical shear profiles measured by probes on P1 to P7, when sparging through gas diffuser E

Gas flow rate: 195 mL/min; gas nozzle size: 3 mm; bundle configuration: tightly-held

Similar to the results for the diffuser at location A (Figure 7-3), test-fibers P3 to P7 were shielded by test-fibers P1 and P2. Interestingly, the measured signals for test-fibers P3 and P7 were similar for diffuser locations A and E. These results suggest that the rising bubbles induced bulk liquid flow throughout the cylindrical tank, and that this bulk flow was not substantially impacted by the nozzle locations investigated.

7.3.1.2 Effect of Gas Nozzle Location

Sparging close to bundle through Nozzle A

The shielding effect described in Section 7.3.1.1 is presented in Figure 7-5a, which shows the time-average surface shear signals from test-fibers P1 to P7 during gas sparging through gas nozzle A. Each bar on the figure represents the time-averaged surface shear signal of one fiber, arranged in the same configuration seen in Figure 7-1. Test-fibers P1 and P2, which were the test-fibers closest to the gas nozzle, recorded the highest time-averaged surface shear signal. Test-fibers P3 to P7, which were located behind test-fibers P1 and P2 recorded lower timeaveraged surface shear signals than test-fibers P1 and P2. Although the differences in surface shear signals between the test-fibers immediately adjacent to the nozzle (i.e. test-fiber P1) and test-fibers not adjacent to the nozzle (i.e. P3) may not seem very significant, a small difference in the value of surface shear signal actually results in a significant difference in the value of shear stress. This is due to the fact that shear stress (Pa) calculated from the surface shear signal (V) is raised to the power 3 (see Equation 4-14). This is illustrated in Figure 7-5c and d, where the shear stresses were estimated based on a shear signal voltages using Equation 4-14. The shear stress experienced by test-fibers P1 and P2 was 4 to 5 times greater than that experienced by testfibers P3 to P7. This observation is consistent with results from a particle image velocimetry study of axial velocities inside a 3 x 3 fiber bundle – where axial velocities outside the hollow fiber bundle were reported to be up to 10 times higher than the velocities inside the fiber bundle [109].



Figure 7-5. Measured time-averaged surface shear signals and calculated shear stresses (Pa) acting on P1 to P7 during gas sparging through nozzle A

(a) Measured time-averaged surface shear signal (V) -1 mm nozzle, (b) Measured time-averaged surface shear signal (V) -3 mm nozzle, (c) Calculated time-averaged shear stresses (Pa) -1 mm nozzle, (d) Calculated time-averaged shear stresses (Pa) -3 mm nozzle. Gas flow: 195 mL/min; fiber configuration: tightly-held. Shear values are based on the average of two replicate experiments

The shielding of test-fibers P3 to P7 might result in more extensive fouling in these fibers during filtration. Yeo and Fane [135] found that fibers within a fiber bundle fouled more quickly, compared to a single fiber under the same experimental conditions. The higher fouling rate was attributed to "stagnant" flow areas within a bundle. This "stagnant" area or shielding effect can potentially lead to inter-fiber fouling, where cake growth on individual fibers begins to merge with the cake layers on other fibers, resulting in several fibers binding to each other through the

cake layer [134, 135]. The binding can further enhance the shielding effect within the bundle and become even more detrimental to fouling control [135].

It is possible that increasing the gap width between fibers (decreased packing density) will reduce the shielding effect, and decreasing the gap width (increased packing density) will increase the shielding effect. Chang and Fane [65] suggested that high fiber packing density results in less favorable hydrodynamic conditions inside the bundle. However, decreased packing density reduces the available surface area for filtration and can increase the cost of a membrane system. An optimum packing density, which balances the hydrodynamic and economic factors, likely exists.



Figure 7-6. Standard deviation of time-averaged surface shear signals and calculated shear stresses acting on P1 to P7 during gas sparging through nozzle A

(a) Standard deviation of time-averaged surface shear signal (V)– 1 mm diffuser, (b) Standard deviation of time-averaged surface shear signal (V)– 3 mm nozzle, (c) Standard deviation of the calculated time-averaged shear stresses (Pa) – 1 mm nozzle, (d) Standard deviation of the calculated time-averaged shear stresses (Pa) – 3 mm nozzle. Gas flow: 195 mL/min; fiber configuration: tightly-held. Values are based on the average of two replicate experiments



Figure 7-7. Total number of shear events of P1 to P7 during gas sparging through nozzle A (a) 1 mm nozzle, (b) 3 mm nozzle

Sparging farther away from fiber bundle- nozzles B, D and E

The time-averages and standard deviations of surface shear signals for test-fibers P1 to P7 when sparging occurred through nozzles B, D and E are presented in Figure 7-8, Figure 7-9 and Figure 7-10, respectively. Nozzles B and E were located farther away from the bundle than nozzle A. As a result, time-averages and standard deviations of surface shear signals experienced by the fibers were lower than those observed in for nozzle A. The total number of shear events measured by test-fibers P1 to P7 when sparged through nozzles B, D and E were also substantially less than those sparged through nozzle A. The total number of shear events of the fibers, sparged through nozzles B, D and E at different gas flow rates are presented in Appendix J. When gas sparging through nozzle D, which was close to test-fibers P1 and P2, but off-center, the bubbles were observed to rise at an angle towards the left of test-fiber P1 rather than vertically. As a result, the distance between test-fibers P1 and P2 and the rising gas bubbles were relatively large, resulting in relatively low time-averages and standard deviations of surface shear signals. These results indicate that the distance between the fibers and the gas bubbles also affects the magnitude and variability of shear events.

Figure 7-11 shows a summary of time-average surface shear signals of test-fibers P1 to P7 for different gas nozzles A, B, D and E. As discussed earlier, sparging through nozzle A resulted in the highest time-average surface shear signals, and this is especially true for test-fibers P1 and P2. Figure 7-11 also shows that the surface shear signals of test-fibers P3 to P7 were lower than those of test-fibers P1 and P2. These results indicate the shielding effect, discussed earlier.



Figure 7-8. Average and standard deviation of surface surface shear signals acting on P1to P7 when sparging through nozzles B

(a) Calculated average surface shear signal -3 mm nozzle, (b) Calculated average surface shear signal -1 mm nozzle, (c) Calculated standard deviation of surface shear signal -3 mm nozzle, (d) Calculated standard deviation of surface shear signal -1 mm nozzle. Gas flow: 195 mL/min; fiber configuration: tightly-held



Figure 7-9. Average surface shear signals and standard deviation of surface surface shear signals acting on P1to P7 when sparging through nozzles D

(a) Calculated average surface shear signal -3 mm nozzle, (b) Calculated average surface shear signal -1 mm nozzle, (c) Calculated standard deviation of surface shear signal -3 mm nozzle, (d) Calculated standard deviation of surface shear signal -1 mm nozzle. Gas flow: 195 mL/min; fiber configuration: tightly-held



Figure 7-10. Average surface shear signals and standard deviation of surface shear signals acting on P1to P7 when sparging through nozzles E

(a) Calculated average surface shear signal -3 mm nozzle, (b) Calculated average surface shear signal -1 mm nozzle, (c) Calculated standard deviation of surface shear signal -3 mm nozzle, (d) Calculated standard deviation of surface shear signal -1 mm nozzle, Gas flow: 195 mL/min; fiber configuration: tightly-held



Figure 7-11. Average surface shear signals of P1 to P6 for different gas nozzles

a) gas flow rate 95 mL/min, b) gas flow rate 142 mL/min, c) gas flow rate 195 mL/min

7.3.1.3 Effect of Gas Sparging Rate

Figure 7-12 shows the time-averaged surface shear signal at different gas sparging rates for all fibers (test-fibers P1 to P7) when gas sparging through nozzle A. For test-fibers P1 and P2, which were closest to gas nozzle A (test-fibers P1 and P2), increasing gas sparging rates resulted

(b)

•P1

P2

▲P3

AP4

oP5

□P6

ł

T

Е

В

Nozzle

D

in increased time-averaged shear. For test-fibers P3 to P7, which were shielded by the test-fibers P1 and P2, increasing gas sparging rate from 10 to 95 mL/min also resulted in increased averaged surface shear signal. However, beyond the gas sparging rate of 95 mL/min, further increases in sparging rate did not result in increased surface shear signals. This trend was observed regardless of the diffuser size (1 mm vs 3 mm). This is consistent with the observation by others that for a gas-sparged submerged hollow fiber membrane module, a critical gas flow rate exists above which no further improvement in fouling control can be achieved[71, 181].



Figure 7-12. Time averaged surface shear signal at different gas sparging rates for all fibers (P1 to P7) when gas sparging through nozzle A

(a) 1 mm nozzle (b) 3 mm nozzle, Fiber configuration: tightly-held

The trends observed for sparging through nozzle A for different gas flow rate were not as evident as those observed for sparging through nozzles B, D and E, as shown in Figure 7-13 (for Nozzle D). The difference was likely due to the fact that for nozzles B, D and E, the gas bubbles were too far away from the fibers to have an impact of inducing high shear stresses on the fibers, compared to when sparging through nozzle A. Additionally, it is possible that, although a higher gas flow rate would increase bulk mixing, it however would not increase bulk flow near the fibers, as the system tank does not have a riser and downcomer section. A riser and a downcomer section in the tank would facilitate an increased bulk flow, as reported by Prieske et al.[133]. However, as discussed in Section 5.3, bulk liquid flow does not significantly contribute

to the overall surface shear signal compared to the surface shear signal induced by the gas bubbles.



Figure 7-13. Time averaged surface shear signal at different gas sparging rates for all fibers (P1 to P7) when gas sparging through nozzle D. The trends are similar for nozzles B and E

(a) 1 mm nozzle (b) 3 mm nozzle, Fiber configuration: tightly-held

7.3.1.4 Effect of Diffuser Nozzle Size

As shown in Figure 7-14 and Figure 7-15, increasing gas sparging rates resulted in increased time-average and standard deviation surface shear signals for both nozzle sizes – 1 mm and 3 mm (although the differences between the averages and standard deviations of surface shear signals of the 1 mm and 3 mm nozzles were not statistically significant, this trend was nonetheless consistently observed). Figure 7-16 shows the total number of shear events for gas flow rates of 95, 142 and 195 mL/min, for test-fibers P1 and P2 using the 1 mm and 3 mm gas nozzles. Overall, the total number of shear events for all experiments increased when gas flow rate was increased from 95 to 142 mL/min. There was no consistently substantial difference between the total number of shear events for gas flow rates of 142 and 195 mL/min. There was no consistently substantial difference between the 1 mm and 3 mm nozzles. These observation contradicts the previous observations reported in Section 5.3 and 6.3 which indicated that a higher total number of shear events were generated for smaller gas nozzle sizes, for a

given flow rate. It is noted that in the system used in Section 5.3 and 6.3, the bubbles were relatively confined within a bundle of fibers, whereas the results presented here are for bubbles that were introduced at the periphery of the fiber bundles. In addition, the system investigated in Section 5.3 and 6.3 used a lower gas flow rate (10 to 35 mL/min). This highlights the importance of membrane configuration and gas sparger design in determining the effect of bubble size on fouling, and likely explains why contradictory results are reported in literature with regards to the effect of bubble size on fouling control.



Figure 7-14. Comparison of average shear stresses at different gas sparging rates when sparging through the 1 mm and 3 mm gas nozzle (at Nozzle A)

(a) P1, (b) P2. Error bar correspond to confidence interval equivalent to one standard deviation of replicate experiments



Figure 7-15.Comparison of the standard deviation of surface shear signal at different gas sparging rates when sparging through the 1 mm and 3 mm gas nozzle (at Nozzle A)

(a) P1, (b) P2. Error bar correspond to confidence interval equivalent to one standard deviation of replicate experiments



Figure 7-16. Total number of shear events for gas flow rates 95, 142 and 195 mL/min, for P1 and P2 and using the 1 mm and 3 mm gas nozzles

Error bar correspond to +/- one standard deviation of replicate experiments

7.3.1.5 Effect of Fiber Tension

In submerged hollow fiber modules, a certain degree of fiber looseness has been found to yield enhanced permeate flux, compared to tightly-held fibers [10, 11, 86, 142]. In a bundle of loosely-held fibers, the gaps between the fibers are more random. As a result, the probability of a gas bubble entering the bundle and imparting high shear stresses on interior fibers is higher, compared to a bundle of tightly-held fibers. Figure 7-17. shows the shear distribution of loosely-held fiber under the same gas sparging rate, and nozzle location as that presented in Figure 7-5. The shielding effect observed for tightly-held bundles (Figure 7-5) is not as pronounced as that observed for loosely-held bundles (Figure 7-17 and Figure 7-8). There is also a more even distribution of shear within the fiber bundle, which will likely result in better fouling control.



Figure 7-17. Average surface shear signals acting on P1 to P7, loosely-held

Nozzle A. a) 1 mm gas diffuser, b) 3 mm gas diffuser. Gas flow rate - 195 mL/min

7.3.1.6 Effect of Fiber Movement

In addition to better shear distribution due to increased gap width between fibers, loosely-held fibers may also offer the benefit of increased shear stresses within the bundle due to fiber movement. Figure 7-18 shows the effect of fiber movement on shear distribution within the fiber bundle as observed in this study. Results from the application of two different frequencies of fiber movement are shown. For both movement frequencies, a more even distribution of surface

shear signal within the bundle was observed, and faster fiber movement resulted in higher surface shear signals. The benefit of fiber movement is clearly the result of inducing a better distribution of shear stresses within the bundle. It is noted here that to date, there is no known comprehensive study on the effect of fiber movement on shear stress distribution in a full-scale submerged hollow fiber module under gas sparging, due to the complexity of hydrodynamic conditions that exists for such systems. Studies by several authors on the benefits of loosely-held moving fibers were based on bench-scale or pilot scale systems. Similarly, the experiments in the present study were also conducted at bench-scale. It is noted here that the extent of fiber movement is expected to be related to fiber length. Since fibers in bench scale systems may not be as pronounced. Despite this, however, the present study provides an insight into the beneficial effect of fiber movement from the point of view of hydrodynamics – shear stresses, which has not been demonstrated before.



Figure 7-18. Effect of fiber movement on shear distribution within the fiber bundle

a) 1 cycle per sec 2 cm amplitude, b) 1 cycle per sec 4 cm amplitude
7.3.1.7 Effect of Physical Contact between Fibers

Figure 7-19 shows the shear profile on test-fiber P1 when physical contact was simulated by touching the test-fiber P1 with another fiber while sparging at a rate of 35 mL/min, and through nozzle A. During fiber contact, the magnitudes of the peaks in shear signal were much higher than those observed without physical contact. Physical contact between fibers was simulated three times, and the associated peaks were labeled 1, 2 and 3 on Figure 7-19. A spike in shear signal was observed during physical contact. Although the principal of the electrochemical shear probe may not be valid during physical contact at the shear probe surface, an indication of increased peak in shear signal during physical contact suggests an increase in the magnitude of shear stress. As such, fouling control is likely to be greatly enhanced in systems in which physical contact between fibers is encouraged. The benefits of physical contact between fibers, however, may be counter balanced by the increased weathering of fibers during membrane operation.



Figure 7-19. Shear profile of loosely held test-fiber P1 with physical contact Contact was simulated three times, labeled 1, 2 and 3 on the shear profile. Gas sparging rate was 35 mL/min

7.4 Conclusions

A mapping of the shear distribution on the surfaces of a bench-scale hollow fiber bundle under different gas sparging conditions was performed. The effect of gas nozzle size, nozzle location, gas sparging rate, fiber configuration, as well as fiber movement and physical contact between fibers were examined. The following are the main conclusions from the present study.

- 1. A significant shielding effect was observed in tightly held fibers- whereby test-fibers located within the bundle experienced substantially lower magnitudes and variability in surface shear signal than the fibers at the peripherals and adjacent to the gas nozzle.
- Increasing gas flow rate resulted in increased magnitudes of the measured surface shear signal. However, there was a critical gas flow rate, beyond which any further increases in gas flow rate did not contribute to higher shear experienced by the test-fibers.
- 3. The shielding effects observed in bundles with tightly-held fibers were lower with loosely-held fibers. For loosely-held fibers, the surface shear signals were more homogeneously distributed within the fiber bundle. Fiber movement was observed to induce shear stresses.

8. Relationship between Types of Shear Profiles and Membrane Fouling

8.1 Introduction

Membrane fouling occurs when material accumulates on a membrane and forms a concentration polarization layer and/or cake layer on the surface, or when it completely or partially blocking the pores of the membrane [42]. These mechanisms increase the resistance to permeate flow. Material accumulation and the formation of a concentration polarization or cake layer is largely dependent on the suspension composition, membrane properties and operating conditions [163]. Operating conditions, such as the hydrodynamic conditions near the membrane play a very important role in the erosion of the cake layer, thus improving overall membrane permeability.

Shear stress has been recognized as an important parameter in controlling particle back-transport from membrane surfaces, by eroding the cake layer [10, 94]. Several mechanistic models have been developed to describe particle back-transport from membrane surfaces, e.g. the shear-induced diffusion, the inertial lift and the surface transport models [3]. However, these models assume constant, laminar flow conditions, such as those observed during single-phase crossflow inside confined membrane systems (e.g. tubular membranes) [3]. On the other hand, the hydrodynamic conditions inside submerged gas-sparged hollow fiber membrane systems are characterized by non-repeating oscillating flows which generate highly variable and transient shear conditions at membrane surfaces [107, 183]. Ochoa et al. [111] found that non-uniform, variable shear stress imposed near the biofilm is much more effective in detaching the biofilm than a constant shear flow imposed on the membrane surface. These observations suggest that non-uniform, transient shear stress plays an important role in the removal of foulants on membrane surfaces.

There have been several attempts to experimentally establish a relationship between the transient shear stress and fouling control in oscillating flow conditions. Several statistical shear parameters such as the mean shear stress, total shear stress, amplitude of shear stress, and the frequency of oscillation were considered to attempt to establish relationships [47, 92, 93, 143, 144]. Although these relationships highlight the importance of shear stress in controlling fouling and improving overall filterability, the proposed relationships between the shear parameters and fouling control do not provide insights into the mechanisms of fouling control.

The motivation for the work described in the present chapter began with the observations that there are different types of shear profiles near the fiber surface in a submerged gas sparged hollow fiber membrane system, due to differences in bubble dynamics that occur in the water matrix [183]. Bubble dynamics are affected by membrane configuration, including packing density, gas sparger design, degree of fiber looseness, and flow properties (i.e. gas and liquid flow rates, matrix viscosity). Three different bubble dynamics scenarios, with respect to the distance between the bubbles and the fiber surface, were observed: (1) bubbles in contact with the fiber, (2) bubbles in close vicinity to the fiber but not in contact with the fiber, and (3) bubbles far away from the fiber. As previously discussed in section 5.3.1.2, a bubble in contact with a fiber results in very high transient shear stresses of short duration at the fiber. These shear stresses are generated by the tail end of the bubble due to secondary flows. Bubbles rising far away from the membrane surface result in relatively constant shear stress due to the entrained liquid flow (i.e. single-phase flow).

For bubbles rising near a fiber but not in contact with the fiber, different types of shear profiles can be observed. Figure 8-1 shows two typical shear profiles observed during the passage of one bubble in a bench-scale gas sparged submerged hollow fiber module. One of the profiles exhibits a shear peak with a relatively short duration, while the other exhibits a shear peak that is similar in magnitude, but with a longer duration. The profiles are repeated during the passage of several gas bubbles. Depending on the magnitude as well as the frequency of these repeating shear profiles, one may obtain similar statistical shear parameters (e.g. time averaged values and amplitudes (maximum and minimum) of shear stress) for both the profiles presented in Figure 8-1a and Figure 8-1b, even though these profiles are different. To date, it is not known whether the different shear profiles generated by these different scenarios result in different mechanisms and/or degrees of fouling control during filtration.



Figure 8-1. Two typical shear profiles observed in a gas sparged submerged hollow fiber module. The bubble is not in contact with the fiber

a) High shear with long duration, b) high shear with relatively shorter duration. Magnitudes of peak shear for both cases are lower than the peak shear when bubble is in contact with the fiber

The overall objective of the present study was to *qualitatively* identify the types of shear profiles that produce the greatest beneficial effect on minimizing reversible surface fouling. The relationship between the different statistical shear parameters that have been used by others to establish a relationship and fouling control (e.g. time averaged shear, standard deviation of shear and amplitude of shear) were examined as well. A number of shear profiles of different magnitudes, durations and frequencies were chosen to simulate the three bubble scenarios, with respect to the distance between the bubbles and the fiber. Filtration experiments were performed under these simulated shear scenarios.

8.2 Experimental Methods

Four different types of shear profiles with varying frequencies were simulated.

- 1. **Single-phase shear profile (Single-phase)** sustained shear stresses with limited variation in shear (i.e. non-transient shear), similar to the shear profiles during single-phase liquid flow with no gas sparging.
- Sustained peak shear profile (SP) transient, lower shear stresses generated when bubble is close to fiber, but not in contact with fiber (Figure 8-1a). Peak shear stress is of longer duration than the HP and the LP shear profile.
- Low peak shear profile (LP)- transient, lower shear stresses generated when bubble is close to fiber, but not in contact with fiber (Figure 8-1b). Peak shear stress is of short duration.
- 4. **High peak shear profile (HP)** transient, high shear stresses generated when bubble comes in contact with fiber. Peak shear stress is of short duration

The fouling rate during filtration was assessed by monitoring the increase in suction pressure over time when the membrane was subjected to the different types and frequencies of shear profiles when filtering solution containing bentonite clay particles. A faster pressure increase was taken to indicate a greater fouling rate. To ensure that only reversible fouling (i.e. cake formation) on the membrane occurred, bentonite particles larger than the membrane pore size were used. This eliminated the possibility of irreversible fouling of the membrane during filtration.

8.2.1 Description of the Bench-Scale Filtration Apparatus

The filtration apparatus consisted of a shear apparatus, a test-fiber, a hollow fiber membrane, a permeate flux pump, and a pressure monitoring system. The shear apparatus (discussed below) was used to create several repeatable shear profiles during the filtration experiments using the test-fiber. For the filtration experiments, a hollow fiber membrane was used.

The shear apparatus consisted of a cylindrical tank (i.d.19 cm), an impeller system, a rig which secured the impeller system, and a test-fiber on which an electrochemical shear probe was embedded, as shown in Figure 8-2. The impeller system consisted of a motor and different types of impellers which were capable of generating shear profiles that were similar to those induced by gas sparging in submerged hollow fiber membrane systems (seen in Figure 8-1). Different shear profiles with controllable magnitudes, duration and frequencies were generated by changing the impeller blade geometry, the distance between the impeller blade and the test-fiber, and the impeller rotation speeds. A total of 4 different types of shear profiles listed below, were generated using this system:

- i. the single-phase shear profile,
- ii. the sustained peak shear profile,
- iii. the low peak shear profile,
- iv. the high peak shear profile.

The sustained, low peak and high peak shear profiles provided repeating transient shear stresses similar to those seen in Figure 8-1a and b, while the single shear profile simulated sustained non-transient shear stresses, similar to that observed inside a confined tube subjected to single-phase crossflow. The blades used for generating these types of shear profiles are shown in Figure 8-3. The relative position of the different impeller blades to the fiber is shown in Figure 8-4. For the low peak shear profile, the impeller blade was raised 2 cm above the fiber. For the high peak shear profile, there was no visually observable gap between the impeller blade and the fiber. As such, it is possible that there was contact between the impeller blade and the fiber. For the sustained peak and the single-phase profile, the distance between the impeller blade and the test-fiber/hollow fiber was small, leaving a gap of approximately 0.5 mm between the fiber and the blade.



Figure 8-2. Picture of the shear apparatus

a) Side view, b) Top view



Figure 8-3. Typical impeller blades of the shear apparatus

a) Single-phase, b) Sustained peak with 2 blades, c) Sustained peak with 4 blades, d) Low peak and high peak with 2 blades, e) Low peak and high peak 4 blades,



Figure 8-4. The side view of tank that shows the relative position of the different types of impellers to the test fiber

a) Single phase, b) Sustained peak, c) Low peak, d) High peak

The rotational speeds of the impellers were adjusted to achieve comparable shear profiles between each experiment (i.e. comparable time averaged shear stress, peak and baseline shear stresses). For the sustained peak shear profiles – the rotational speed was set at 5 rpm. For the low peak and high peak shear experiments the rotational speed was 8.5 rpm. For the single-phase shear profile, the rotational speed was set at 5 rpm. Additionally, leveling of the shear apparatus was important in ensuring that the shear profiles along the entire length of the fiber were the same (discussed in detail in Appendix K). Careful leveling of the apparatus was conducted prior to the start of the filtration experiments.

The test-fiber, on which a shear probe was embedded on its surface, was used to measure the shear profiles generated during the passage of the impellers. A single probe set up (section 7.2.1) was used to measure all shear profiles, while a double probe set up (section 4.3.2) was used to check the presence of flow reversal near the probe for the sustained peak shear profile. To ensure that the test-fiber was placed securely against the interior wall of the cylindrical tank, the test-fiber was attached onto a piece of flexible rubber backing (length x width = 21.6 cm x 3.8 cm) using epoxy, and placed against the interior wall of the cylindrical tank, as shown in Figure 8-5. Care was also taken to ensure that the placement of the hollow fiber in the filtration apparatus was similar to the placement of the test-fiber, so that during the filtration experiment similar shear profiles to those observed during the shear experiments were obtained.



Figure 8-5. Test-fiber placed inside system tank

(a) Test-fiber on rubber backing, (b) Side view of test-fiber on rubber backing inside system tank, (c) Top view of test-fiber inside system tank, (d) cross-section of the test-fiber and rubber backing

The membrane used for the filtration experiments was a ZW500 type outside-in PVDF hollow fiber membrane, which was provided by GE-Zenon (Oakville, Ontario). The surface properties were non-ionic and hydrophilic, with an outside diameter of 1.8 mm and nominal pore diameter of $0.04 \mu m$.

The total length of the hollow fiber membrane used in the experiment was 20 cm. Similar to the test-fiber, a piece of the flexible rubber was coated with a layer of epoxy, and the hollow fiber membrane was fixed onto the rubber backing. Through visual observations, it was estimated that approximated 40% of the total surface area of the hollow fiber was covered with the epoxy coating, this section of the hollow fiber did not contribute to filtration. The total surface area available for membrane filtration was approximately 1020 mm². The cross-section of the fiber and the rubber backing is shown in Figure 8-5d.

The permeate pump was connected to the fiber membrane and permeate was drawn by suction. The volumetric flow rate of 0.3 mL/min was set to generate a permeate flux of 50 $l/m^2/hr$. This permeate flux is a typical operating permeate flux used for membrane filtration of water through the hollow fiber membrane. A slow flow peristaltic pump (Lachat Instruments Model 2200) was used to generate a constant flow during filtration. The flow was monitored to confirm that it was constant during filtration.

The pressure monitoring system measured and logged the TMP during the filtration experiments. The system consisted of a pressure gauge (Cole Palmer GPI 9675), and a pressure transducer (Omega PX240) connected to a data logger (National Instrument USB-6009). The data logger collected pressure measurements at a rate of 1 Hz. A custom Labview application (Labview Version 7.0) recorded the collected pressure measurements. The signal from the pressure transducer was calibrated using a portable pressure calibrator (DPI 601, Druck Inc.) at the beginning of each experiment (Appendix L).

The only type of fouling to be considered in the filtration experiments was reversible cake fouling. Therefore, the water matrix used for the experiments could not result in irreversible fouling, such as pore clogging, or adsorption onto membrane. A solution containing reverse osmosis (R.O.) filtered tap water and sodium bentonite particles were chosen as the water matrix for the filtration experiments. The average mean size of the bentonite particles was 3 μ m; while the smallest particle size was 0.3 μ m. Since the nominal pore size of the membrane (0.04 μ m) is smaller than the smallest bentonite particles, the only type of membrane fouling that was expected during filtration was reversible cake fouling on the membrane surface. The particle

size distributions of the bentonite solution were analyzed using a laser particle size analyzer (Mastersizer Hydro 2000S, Malvern), and the results are shown in Appendix M. Two bentonite solution concentrations were considered during the experiment: 0.2 and 0.5 g/L.

All experiments were conducted at room temperature. Prior to the start of the filtration experiments, the water was allowed to equilibriate with room temperature ($20 \, ^{\circ}C \, +/-2 \, ^{\circ}C$). A 2 $^{\circ}C$ change in temperature was not expected to affect the filtration pressure, since it only results in a 0.4 % change in the solution viscosity. The duration of filtration experiments with 0.2 g/L bentonite was 120 minutes; the duration of filtration experiments with 0.5 g/L bentonite was 70 minutes. This ensured that suction pressure did not exceed 68.95 kPa (10 psi), which is the manufacturer-recommended maximum operating pressure for the membrane. During filtration, permeate was returned to the system tank to maintain a constant bentonite concentration during filtration.

After each filtration experiment, the hollow fiber membrane was cleaned thoroughly to ensure that all cake foulants formed on the membrane surface during filtration were removed. For cleaning, the membrane was removed from the cylindrical tank and rinsed with R.O.-filtered tap water. The membrane was then placed in a the cleaning apparatus filled with R.O.-filtered tap water and subjected to vigorous gas sparging for 2 minutes. The cleaning apparatus consisted of a long cylindrical column (diameter 3 cm), with an air line connected to the bottom of the column, shown in Figure 8-6. The diameter of the column was small enough such that the membrane rubber backing fitted snugly inside the column, and the bubbles rising in the column were confined between the membrane and the column wall, thus providing effective scouring of the membrane surface. After rigorous gas sparging, the solution in the column was discarded and replaced with fresh R.O.-filtered water, and vigorous sparging was repeated twice. The membrane was then placed in 1 liter of fresh R.O. filtered water, and the clean water pressure at the flux of 50 LMH was measured. The pressure acquired at this flux was recorded and compared to other clean water pressures to ensure that the membrane had been thoroughly cleaned. To ensure that all cake foulants formed on the membrane surface were removed prior to the start of a filtration experiment, and that the membrane recovered all of its permeability, the clean water flux was initially determined after each cleaning cycle. However, it was found that

the clean water suction pressure did not change significantly from one experiment to another. Therefore, clean water flux was subsequently determined only after every 5th or 6th cleaning cycle (see Appendix N). At the end of the day after all experiments were completed, the membranes were cleaned (according to the cleaning procedure above) and submerged overnight in a 50 ppm chlorine solution (using R.O.-filtered water). When membranes were to be used again the next day, they were cleaned with water before the start of experiments.



Figure 8-6. Membrane cleaning apparatus

The membrane was subjected to integrity testing at the beginning and the end of each filtration experiment (before cleaning) to ensure that the membrane was not breached during filtration. The integrity testing apparatus is shown in Figure 8-7. Integrity testing was conducted using a pressure hold test, wherein the membrane was submerged in water and pressurized to 68.95 kPa (10 psi), isolated and the pressure was monitored over a period of 60 seconds. If a pressure decrease greater than 15 % was recorded, then the membrane was considered breached. Additionally, if bubbles were observed leaking from the membrane during the pressure hold test — the membrane was also considered breached.

breaches occurred at the connection point between the membrane and the suction tubing. These breaches most likely occurred during the handling of the membrane during filtration and cleaning. Breached connections were repaired by applying a layer of epoxy on the connections. Integrity tests were performed again after repairing to ensure the breached connection had been repaired.

No breaches were observed along the membrane surface. This indicates that, although there was no visually observable gap between the impeller blades and the fiber for some experimental conditions (i.e. high peak shear profile), there was no significant direct contact between the impeller blades and the fiber. Had there been significant direct contact, breaches in the membrane integrity would have been expected.



Figure 8-7. Membrane integrity testing apparatus

8.2.2 Experimental Program

Four types of shear profiles were considered: the LP shear profile, the HP shear profile, the SP shear profile and the single-phase shear profile. The shear experiments were first performed

using the test-fiber and different type of impellers. In these experiments, it was first determined that repeatable shear profiles could be achieved with the different types of impellers. Shear signals in Volts are presented instead of shear stress. Filtration experiments were then performed under the same shear conditions. Table 8-1 summarizes the experiments conducted in this study. Each experiment was either repeated two times or three times. All experiments, including the replicate experiments were performed in random order.

Bentonite Concentration (g/L)	Shear Type	# Blades	Impeller Rotation Speed (rpm)
0.2	sustained peak	1	5
		2	5
		3	5
		4	5
	low peak	1	8.5
		2	8.5
		4	8.5
	high peak	1	8.5
		2	8.5
		4	8.5
	single-phase		5
0.5	sustained peak	1	5
		2	5
		3	5
		4	5
	low peak	1	8.5
		2	8.5
		4	8.5
	high peak	1	8.5
		2	8.5
		4	8.5
	single-phase		5

Table 8-1. Experimental program

8.3 Results and Discussions

8.3.1 Typical Shear Profiles

The four different types of shear profiles that were generated in the filtration experiments are presented below.

Single-phase Shear Profile

Single-phase shear profiles were characterized by a constant surface shear signal with limited variability in shear stresses. The single-phase shear profile was created using a disc as presented in Figure 8-3a. The shear profile created is shown in Figure 8-8 where the surface shear signal is relatively constant at approximately 0.5 V.



Figure 8-8. Single-phase shear profile

Sustained Peak Shear Profiles

The sustained peak shear profiles were created using the sustained peak impellers (seen in Figure 8-3b and c). The shear profiles, shown in Figure 8-9 a to d consist of peak surface shear signals (around 0.5 V) of relatively long duration $(3-5 \text{ seconds})^7$. Shear profiles with different frequencies were created with the different number of impeller blades. Replicate experiments (a,

 $^{^{7}}$ The duration of the peak observed in Figure 8-8(3 - 5 s) is different than the duration of the peak presented in Figure 8-1b (0.2 s). However, their shear profiles are very similar.

b and c) are shown in each figure. It can be seen that the repeatability of the shear profiles was good.

The sustained peak shear profiles in Figure 8-9 are similar to those seen in earlier observations of typical profiles in a gas sparged system (Figure 8-1a), wherein the amplitudes of the profiles were of the same order of magnitude as that seen in Figure 8-1a. The peak shear values in Figure 8-1a were measured during the passage of a rising gas bubble. Similarly, the peak surface shear signals seen in Figure 8-9 were measured during the passage of the impeller blades. During the brief period of the absence of the blade, there was some residual movement of the entrained liquid in the direction of the rotating blade. A larger number of blades on the impeller resulted in increased velocity of the entrained liquid. As a result, the baseline for the 4 blade shear experiment were higher (0.35 V) compared to the 3 blade experiment (0.28 V), the 2 blade experiment and the 1 blade experiment (0.25 V).

The separation distance between the blade and the shear probe (or hollow fiber membrane) was small (i.e approximately 0.5 mm). Since the blades were very close to the shear probe (or hollow fiber membrane), there was a possibility that the flows near the probes may have been confined by the block blades, similar to the situation of a slug flow rising in a confined tube. This may have resulted in flow reversal near the membrane surface. A different set of experiments, using the double-probe mounted flush on the test-fiber was conducted to check whether flow reversal occurred near the membrane during the passage of the blades. Those measurements showed that no flow reversal occurred near the fiber. These results are shown in Appendix O.



Figure 8-9. Sustained peak shear profiles: different frequencies created using different number of blades on each impeller

a)– 1 blade, b) – 2 blade, c) – 3 blade, d) – 4 blade

Low Peak Shear Profile

The low peak shear profiles were created using the low peak shear impellers (seen in Figure 8-3d and e). Different frequencies of the low peak shear profiles were created with impellers with

different numbers of blades, shown in Figure 8-10. Similar to the sustained peak shear profiles, the peak and minimum shear values were around 0.5 and 0.3 V, respectively. However, unlike the sustained peak shear profiles, the durations of the peak surface shear signals were relatively short (1 second). The low peak shear profiles are similar to those seen in earlier observations of typical profiles in gas sparged system (Figure 8-1b).

High Peak Shear Profile

The high peak shear profiles were created using the high peak shear impellers (seen in Figure 8-3d and e). Different frequencies of shear events were created with impellers with different numbers of blades, shown in

Figure 8-11. The high peak shear profiles are similar to those of the low peak shear profiles, except that the peak values (1.0 V to 1.6V) are much higher than those seen for the low shear profiles (0.5 to 0.6 V). The minimum surface shear signals were similar to those of the low peak shear profiles (approximately 0.3 V)



Figure 8-10. Low peak shear profiles: different frequencies created using different number of blades on each impeller

a) -1 blade, b) -2 blade, c) -4 blade



Figure 8-11. High peak shear profiles: different frequencies created using different number of blades on each impeller

a) -1 blade, b) -2 blade, c) -4 blade

8.3.2 Typical Filtration Pressure Curves

All of the filtration TMP curves shown in this section are the average pressures calculated from the replicated experiments. The confidence intervals presented in some of the pressure curves correspond to a confidence interval equivalent to one standard deviation of the measured pressure at a given time for the replicated experiments. All statistical comparisons of the pressure curves between the different experiments are based on this confidence interval. For the purpose of presentation, the confidence levels are only shown in the figures after 30 minutes of data for the 0.2 g/L experiment, and after 60 minutes for the 0.5 g/L. Below these times the confidence intervals of the pressure curves overlapped.

8.3.2.1 Pressure Curve for the Single-Phase Shear Experiment

Typical TMP curves for the single-phase shear experiment are shown in Figure 8-12. For both curves (the 0.2 g/L and 0.5 g/L experiments) the TMP increase in the first 5 minutes was rapid, after which the TMP increased slowed over the duration of the experiment. The rapid change during the first five minutes was due to the pressure build up to a level equivalent to that of the clean water filtration pressure. As expected, due to differences in the rates of convective mass transfer, the rate of fouling for the experiment with the higher bentonite concentration (0.5 g/L) was greater than that for the experiment with lower bentonite concentration (0.2 g/L). In the present experiments, the time to reach 68.95 kPa (10 psi) was less than 65 minutes for the 0.5 g/L experiment, while for the time to reach the same pressure was approximately 110 minutes for the 0.2 g/L experiment. This indicates a higher fouling rate in the 0.5 g/L experiment.



Figure 8-12. Fouling for single-phase shear profile

Upper and lower limit corresponds to a confidence interval equivalent to one standard deviation of the measured pressure at a given time for the replicated experiments

8.3.2.2 Pressure Curve for the Sustained Peak Shear Experiment

Typical TMP curves for the sustained peak shear experiments are shown in Figure 8-13. Similar to the observations from single-phase shear profile experiments, the fouling rate for the 0.5 g/L suspension was greater than that for the 0.2 g/L experiments (i.e. approximately double). For the 0.2 g/L experiment, when comparing the effect of different shear frequencies (i.e. number of blades) for each experiment, the 4 blade experiment demonstrated the lowest fouling rate compared to experiments performed using 1, 2 and 3 blades. Unexpectedly, however, the opposite trend was consistently observed for the 0.50 g/L experiment, wherein the 4 blade experiment resulted in a faster fouling rate than the 3 blade and 2 blade experiments. The confidence intervals for the pressure curves of the 3 blade and 4 blade experiments are shown in Figure 8-14. Since the confidence levels do not overlap, the difference in the pressure profiles of the 3 blade and 4 blade experiment and the 0.2 g/L experiment.

There was also no statistical difference between the 1 blade, 2 blade and the 3 blade experiments for the 0.5 g/L and 0.2 g/L experiments. A possible explanation for this observation is that there may be a minimum required energy for fouling control to take place, below which the control of fouling is not effective. In a study of the crossflow microfiltration of skim milk inside a tubular membrane, Leberre et al [94] found that there was a critical shear stress that was required before cake erosion occurred on the membrane surface. Similarly, in the present study, the shear stresses (and indirectly the energy) supplied by the 3 blade shear conditions may be the minimum (or critical) energy required to control fouling for this particular system. Below this critical shear condition (i.e. 1 blade and 2 blade) no difference in fouling control was observed; above this shear condition, an improvement in fouling control was observed (as in the case of the 0.2 g/L experiment). Further research is required to confirm this hypothesis.



Figure 8-13. Fouling for the sustained peak shear profile

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/L



Figure 8-14. Fouling for the sustained peak 3 blade and 4 blade shear profiles with confidence interval

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/L. Upper and lower limit corresponds to a confidence interval equivalent to one standard deviation of the measured pressure at a given time for the replicated experiments

8.3.2.3 Pressure Curve for the Low Peak Shear Experiment

Typical TMP curves for the low peak shear experiments are shown in Figure 8-15 and Figure 8-16. Similar to the observations from the single-phase and sustained peak shear profile experiments, the fouling rates of the experiment with the higher bentonite concentration (0.5 g/L) were higher than those for the 0.2 g/L experiment (i.e. approximately double). For both the 0.5 g/L and 0.2 g/L experiments, the fouling rates for the 1, 2 and 4 blade experiments were relatively similar (i.e. confidence intervals overlapped as presented in Figure 8-16). These results suggest that the frequency of shear events alone is not enough to describe the relationship between shear and fouling.



Figure 8-15. Fouling for the low peak shear profile

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/L



Figure 8-16. Fouling for the 1 blade and 4 blade low peak shear profile with confidence interval

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/L. Upper and lower limit corresponds to a confidence interval equivalent to one standard deviation of the measured pressure at a given time for the replicated experiments

8.3.2.4 Pressure Curve for the High Peak Shear Experiment

Typical TMP curves for the high peak shear experiments are shown in Figure 8-17. Similar to the observations from the single-phase, block and low peak shear profile experiments, the fouling rates of the experiment with the higher bentonite concentration (0.5 g/L) was higher than those for the 0.2 g/L experiment (i.e. approximately double). For both the 0.5 g/L and 0.2 g/L experiments, the fouling rates for the 1, 2 and 4 blade experiments were relatively similar (i.e. confidence intervals overlapped as presented in Figure 8-18). Again, these results suggest that the frequency of shear events alone is not enough to describe the relationship between shear and fouling.



Figure 8-17. Fouling for the high peak shear profile

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/L



Figure 8-18. Fouling for the high peak shear profile with confidence intervals

(a) Bentonite concentration 0.2 g/L, (b) bentonite concentration 0.5 g/LUpper and lower limit corresponds to a confidence interval equivalent to one standard deviation of the measured pressure at a given time for the replicated experiments

8.3.3 Comparison of Filtration Pressure Curves of All Shear Profiles

In this section, the TMP curves of all shear profiles are compared. Figure 8-19 shows the comparison of the pressure curves for a solution containing bentonite at a concentration of 0.2 g/L, while Figure 8-20 shows the same comparison for a solution containing bentonite at a concentration of 0.5 g/L. For presentation purposes, pressure curves with and without confidence intervals are placed side by side, since on some figures the curves and the confidence intervals overlap.

For the 0.2 g/L experiments, the fouling rates for the transient shear experiments (i.e. the high peak, sustained peak and low peak shear profiles) were less than that for the single-phase shear profile. The confidence intervals of the low peak and single-phase shear experiments overlap. However, for all experiments, the fouling rate for the low peak shear profile was consistently less than that of the single-phase shear profile. As observed for the 0.2 g/L experiment, for the 0.5 g/L experiment, the fouling rate of the high peak and the sustained peak shear profiles was less





Figure 8-19. The comparison of the fouling for different shear profiles with bentonite concentration of 0.2 g/L $\,$

(a, b) 4 Blade, (c, d) 2 blade, (e, f) 1 blade , (a - c) time averaged shear only, (d - f) time averaged shear with confidence interval



Figure 8-20. The comparison of the fouling for different shear profiles with bentonite concentration of 0.5 g/L

(a, b) 4 Blade, (c, d) 2 blade, (e, f) 1 blade, (a - c) time averaged shear only, (d - f) time averaged shear with confidence interval

Figure 8-19 and Figure 8-20 show that, with the exception of the low peak shear profiles, all transient shear profiles resulted in significantly better fouling control during filtration, compared to the single-phase shear profile. This was expected, and is consistent with the observations of others when investigating fouling under single phase and double phase flow conditions [86, 88, 91, 99, 121, 123]. It is however interesting to note that amongst the different transient shear profiles, different extents of fouling were observed. The high peak shear profiles resulted in the least fouling, followed by the sustained peak shear profiles. The low peak shear profile resulted in the worst fouling, compared to the other two transient shear profiles, and was similar to that of single phase shear profile. One possible explanation for this observation is that, as discussed in Section 8.3.2.2, there may be a minimum (or critical) energy required before particle transport away from membrane can occur. Therefore, the energy supplied by the low peak and single phase shear profile may not have been sufficient in inducing the particle transport, compared to the block and high peak shear profile. Additionally, as discussed in Section 8.2, there was no visible gap between the impeller blade and the fiber membrane for the high peak shear condition. As such, it is possible that physical contact occurred with the fiber membrane. This contact may have resulted in the physical scouring of the membrane surface, which disrupted and removed the cake layer formed on the membrane surface.

When comparing the low peak shear conditions to the sustained peak shear profile experiments, the sustained peak shear profile resulted in better fouling control. The greater fouling observed for the low peak shear conditions is possibly due to the disruption of particle back-diffusion due to excessive oscillatory flow. As discussed in Chapter 3, inertial lift, one of the mechanisms that possibly contribute to fouling control can be impacted by oscillatory flow conditions. Under oscillatory flow conditions, there exists a critical oscillation frequency, above which the flow conditions become detrimental to the lift forces that transport particles away from the membrane [110]. Analogously, the same phenomenon may have occurred in the experiments of the present study, wherein the oscillatory flows provided by the sustained peak shear conditions were beneficial in controlling fouling, compared to the non-oscillatory flows of the single-phase shear conditions. However, at a higher frequency of oscillatory flow, such as that of the low peak shear conditions, fouling control was not as efficient, possibly due to increased secondary flows near the membrane surface that were more detrimental to fouling control. Further research is

required to confirm this hypothesis. Additionally, considering that the peak values of the transient shear stress in the sustained peak shear profile and the low peak profile were similar (around 0.5 V), the results suggest that shear stress peak value does not provide a good indicator of the fouling control. Rather, the profile of transient shear, i.e. sustained peak vs low peak is also an important factor that affects the mechanism of fouling.

8.3.4 Relationship Between the Fouling Rate and Different Shear Parameters

Several researchers have proposed that the permeate flux (and subsequent fouling control) can be linked to several statistical shear parameters, as listed in Table 8-2. For the present study, in addition to the statistical shear parameters listed in Table 8-2, other statistical parameters such as number of shear events (N_s) and the duration of the peak shear (T_{max}) were also considered. Several combinations of these statistical parameters were evaluated as potential relationships that could be use to link shear measurements to fouling control. These evaluated parameters are listed in Table 8-3 for the different shear profiles considered.

Shear Parameter	Symbol	References which suggest relationship to fouling
Time-averaged shear	$\overline{ au}$	[92, 93, 143]
Standard deviation of shear profile	${ au}_{\scriptscriptstyle std}$	[184]
Amplitude of shear profile	${ au}_{amp}$	[93]
Peak shear	${ au}_{ m max}$	[144]
Oscillation frequency	f	[93], [47]
Ratio of two-phase time-averaged	$\overline{\tau_{\mathrm{two-phase}}}$	[93]
shear stress to single-phase wall shear	$\frac{\tau}{\tau}$	
	single-phase	

Table 8-2. Suggested statistical shear parameters linked to fouling

Shear Parameter	Symbol
Number of shear events	Ns
Duration of peak shear (in seconds)	T_{max}
Product of number of shear events and	$N_s \ge \overline{\tau}$
average shear	5
Product of number of shear event and	$N_s \ge au_{\max}$
peak shear	
Product of number of shear event,	$N_s \ge \tau_{max} \ge T_{max}$
peak shear and duration of peak shear	max

Table 8-3. Additional statistical shear parameters considered in the present study as potential link to fouling

The calculated values of these shear parameters for the different shear profiles are shown in Table 8-4. The time-averaged shear and standard deviation of shear were calculated based on 60 minutes of data from the shear profiles.

The fouling rates for all experiments were calculated and plotted against the different shear parameters in Table 8-4. Since none of the pressure curves were linear with respect to time, the fouling rate was calculated based on the change of pressure vs. time near the end of filtration, at which point the pressure increase was approximately linear. For the 0.2 g/L experiment, the fouling rate was calculated between 80 and 120 minutes of the experiment, while for the 0.5 g/L experiment, the fouling rate was calculated between 80 and 120 minutes of the experiment, while for the 0.5 g/L experiment, the fouling rate was calculated between 50 and 70 minutes of the experiment. Details of the regression analysis to estimate the fouling rates are presented in Appendix P. The estimated fouling rates plotted against the values of the different shear parameters are presented in Appendix Q. Based on Pearson's correlation analysis, results in Table 8-5 suggest that $\overline{\tau}$ does not provide a satisfactory description of the relationship between shear and fouling, as initially suggested by others [92, 93, 143]. This observation is consistent with those by Yeo et al. [184] who found that $\overline{\tau}$ cannot be used as the sole parameter in defining fouling control during filtration under two-phase flow conditions. Frequency of oscillation (*f*), and number of shear events (*N_s*), two parameters which were speculated to be linked to fouling [131, 183] were also found to be not good descriptor of fouling rate. Similarly, other parameters investigated

relationship between shear and fouling.

 $^{:\}frac{\tau_{\text{two-phase}}}{\tau_{\text{single-phase}}} \text{ , } f \text{ , } N_s \text{ x } \overline{\tau} \text{ , } N_s \text{ x } \tau_{\text{max}} \text{ , and } N_s \text{ x } \tau_{\text{max}} \text{ x } T_{\text{max}} \text{ also cannot be used to define the}$
The shear parameters that were found to be statistically related to fouling rate were τ_{std} , τ_{max} and $au_{\it amp}$. The relationship between $au_{\it std}$ and fouling rate is consistent with the observations by Yeo et al [184] who demonstrated a good correlation between standard deviation and the rate of transmembrane pressure increase during filtration. This correlation initially suggests the importance of high variability of shear stress in controlling fouling. However, a closer look at the data suggests that τ_{std} is likely not the sole factor in affecting fouling. When comparing the τ_{std} value for the sustained peak-4 shear profile ($\tau_{std} = 0.05$) with the τ_{std} value for the low peak-4, low peak-2 and low peak-1 shear profiles ($\tau_{std} = 0.04$, 0.06 and 0.06, respectively), the τ_{std} values are relatively similar. However, the fouling rates of all low peak shear experiments were significantly higher than those of the sustained peak-4 experiments, as shown in Figure 8-19 and Figure 8-20. Therefore, τ_{std} alone cannot fully describe the relationship between hydrodynamics and fouling. It should also be noted here that although high variability in shear (as described by a higher τ_{std}) may be important for fouling control, there may be a limit to the beneficial effect of shear variability on fouling control. In fact, excessive variability or oscillation in flow may create secondary flow conditions near membrane surfaces that may be detrimental to fouling control [110].

The relationship observed between τ_{max} and the fouling rate is consistent with observations by Jaffrin [144]. However, similar with the discussion of τ_{std} above, τ_{max} alone cannot fully explain why the low peak shear profile yielded a higher fouling rate compared to the sustained peak shear profile, even though the τ_{max} of both types of shear profiles were similar. This is similar for τ_{amp} as well, where the amplitudes of shear stress for the low peak and sustained shear profiles were similar, however, the low peak shear profile yielded a higher fouling rate compared to the sustained shear stress of the sustained peak. Therefore, the amplitude, and the maximum shear stress of the transient shear conditions alone cannot be used to fully describe the relationship between hydrodynamics and fouling either.

The results from the present study suggest that inducing different types of shear events may have different physical effects on fouling control at the membrane surface. For example, the mechanism of fouling control in the high peak shear experiment may have been primarily due to scouring of the concentration polarization and/or cake layers, while fouling control in low peak and sustained shear experiments may have resulted primarily from particle back-transport via inertial lift or shear-induced diffusion. These physical effects cannot be properly described by the simple statistical shear parameters proposed in Table 8-2 and Table 8-3. Further research is required to investigate the mechanisms of fouling control induced by the different shear conditions, and to develop a better description of the relationship between fouling control and the different transient shear conditions.

Shear Profile – number of blades	$\overline{ au}$	${ au}_{\it std}$	$ au_{\it amp}$	$ au_{ m max}$	f	$rac{ au_{ ext{two-phase}}}{ au_{ ext{single-phase}}}$	N_s	T _{max}	$N_s \ge \overline{\tau}$	$N_s \ge au_{ m max}$	$N_s \ge au_{\max} \ge T_{max}$
sustained peak-4	0.46	0.05	0.2	0.5	0.28	0.91	1020	2.5	469	510	1275
sustained peak -3	0.42	0.09	0.2	0.5	0.22	0.83	780	2.5	328	390	975
sustained peak -2	0.37	0.1	0.2	0.5	0.15	0.73	540	2.5	200	270	675
sustained peak -1	0.33	0.1	0.2	0.5	0.08	0.65	300	2.5	99	150	375
high peak-4	0.48	0.15	0.9	1.2	0.57	0.95	2040	1	979	2448	2448
high peak -2	0.43	0.13	0.9	1.2	0.28	0.85	1020	1	439	1224	1224
high peak -1	0.36	0.12	0.9	1.2	0.15	0.71	540	1	194	648	648
low peak-4	0.41	0.04	0.2	0.5	0.57	0.81	2040	1.5	836	1020	1530
low peak -2	0.4	0.06	0.2	0.5	0.28	0.79	1020	1.5	408	510	765
low peak -1	0.33	0.06	0.2	0.5	0.15	0.65	540	1.5	178	270	405
single-phase	0.51	0.02	-	0.2	-	-	-	-	-	_	-

Table 8-4. Shear parameters to different experimental conditions

 Table 8-5. Pearson correlation coefficient for different shear parameters and pressure increase rate

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Bentonite Concentration	$\overline{\tau}$	$ au_{\scriptscriptstyle std}$	${ au}_{amp}$	$ au_{ m max}$	f	$\frac{\overline{\tau_{\rm two-phase}}}{\overline{\tau_{\rm single-phase}}}$	N_s	$N_s \ge \overline{\tau}$	$N_s \ge au_{ m max}$	$N_s \ge au_{\max} \ge T_{max}$
0.2 g/L	-0.129	-0.800	-0.620	-0.630	-0.213	-0.129	-0.213	-0.291	-0.577	-0.305
0.5 g/L	0.019	-0.793	-0.709	-0.711	-0.082	0.019	-0.082	-0.147	-0.505	-0.178

8.4 Conclusions

Different types of shear profiles can be observed in a gas-sparged submerged hollow fiber membrane module. The overall objective of the present study was to *qualitatively* identify the types of shear profiles that produce the greatest beneficial effect on minimizing reversible surface fouling. The relationship between the different statistical shear parameters that have been used by others to establish a relationship and fouling control (e.g. time averaged shear, standard deviation of shear and amplitude of shear) were examined as well. A number of shear profiles of different magnitudes, durations and frequencies was chosen to simulate the three bubble scenarios, with respect to the distance between the bubbles and the fiber. Filtration experiments were performed under these simulated shear scenarios. This study is the first of its kind in investigating the relationship between types of shear conditions and fouling. Shear events of different magnitudes, durations and frequencies were imposed onto a submerged hollow fiber membrane, and the resulting increases in trans-membrane pressure were monitored and analyzed. The following are the main conclusions from this study:

- Filtration experiments in which membranes were subjected to transient shear conditions resulted in lower fouling rates, compared to constant shear conditions (i.e. single-phase shear profile).
- The magnitude, duration and frequency of the shear conditions have an impact on the fouling rate of membranes.
- Shear conditions with high peak values (i.e. high peak shear profile) resulted in the best fouling control compared other shear conditions.
- For a given maximum peak shear value, shear conditions with peak values of relatively long duration (sustained peak shear profile) were more effective at

controlling surface fouling than frequent short shear events (low peak shear profile).

No significant correlations between the fouling rate and the values of shear parameters such as τ , N_s, N_s * τ , N_s x τ_{max}, f, and N_s x τ_{max} x T_{max}. were observed. A possible relationship between τ_{std}, τ_{max} and fouling was observed. However, these parameters alone cannot fully explain why the low peak shear condition yielded a higher fouling rate compared to the sustained peak shear condition, even though the τ_{max} of both types of shear conditions were similar.

9.1 Overall Conclusions

The investigations undertaken in this thesis provide one of the most comprehensive studies of hydrodynamic conditions inside the submerged hollow fiber membrane module to date. To achieve the objectives of the thesis, the first electrochemical double-probe manufactured on a non-rigid surface (with the same dimension and flexibility as a hollow fiber) was developed. This allowed for the detailed investigation of the hydrodynamic conditions inside a bench-scale hollow fiber membrane module, and the relationship between hydrodynamics and fouling. The following are the main conclusions from this study.

Hydrodynamic Conditions

- The hydrodynamic conditions in confined tubular membrane systems and unconfined submerged hollow fiber membranes were observed to be different. These results imply that, in contrast to the situation with confined tubular membranes, flux enhancement in gas-sparged submerged hollow fiber membrane systems is likely not achieved through scouring of the fiber surface by a falling film.
- Different operating conditions yielded bubbles with different geometries (i.e. spherical, ellipsoidal or slug-like), and the different bubble geometries yielded different shear profiles.
- In examining the shear profiles at different radial sections of a fiber during gas sparging, it was noted that the sections of the fiber closest to the sparged bubble experienced shear peaks (i.e events) of the highest magnitude. These results indicate that only a small portion of the fibers benefit from the shear peaks induced by rising sparged bubbles.

- A significant shielding effect was observed in tightly held fibers- whereby fibers inside the bundle experienced substantially lower magnitudes and variabilities in surface shear signal.
- The shielding effects observed in bundles with tightly-held fibers were somewhat lower with loosely-held fibers, and with fibers in motion. For loosely-held fibers, the surface shear signals were more homogeneously distributed within the fiber bundle. The flow path of bubbles rising in loosely held fibers was not confined to a specific region, as was the case for tightly held fibers. The larger number of fibers (i.e. regions) that can benefit from sparged bubbles in systems with loosely held fibers likely explains why higher permeate fluxes have been reported in loosely held submerged hollow fiber membrane systems compared to those in tightly held systems.

Relationship Between Fouling and Shear

- Filtration experiments in which membranes were subjected to transient shear conditions resulted in lower fouling rates, compared to constant shear conditions (i.e. single-phase shear profile).
- The magnitude, duration and frequency of the shear conditions have an impact on the fouling rate of membranes, although the degree of impact varies for different shear conditions.
- Shear conditions with high peak values (i.e. high peak shear profile) resulted in the best fouling control compared other shear conditions.
- Shear conditions with peak values of relatively long duration (sustained peak shear profile) were more effective at controlling surface fouling than frequent short shear events (low peak shear profile).
- No significant correlations between fouling and the values of shear parameters such as $\overline{\tau}$, N_s , $N_s * \overline{\tau}$, $N_s \propto \tau_{max}$, f, and $N_s \propto \tau_{max} \propto T_{max}$. were observed. However, possible relationship between τ_{std} , τ_{max} and fouling was observed.

• By analogy for a gas-sparged membrane system, the distance between a rising bubble and the membrane surface may be an important factor in determining the fouling, as a smaller distance may result in the physical disruption of the cake layer on the membrane surface.

9.2 Recommendation for Future Work

Characterizing Hydrodynamic Conditions Inside Submerged Hollow Fiber Membrane Modules

Fiber movement plays an important role in controlling fouling. Several possible mechanisms of fouling control caused by fiber movement during gas sparging are: (1) creation of a more even distribution of flow and shear stresses inside the fiber bundle (as discussed in Chapter 7), (2) "shaking" off accumulated particles on the membrane surface, and (3) promotion physical contact between fibers which results in the physical scouring of the fouling layer formed on membrane surfaces. In addition to fouling control, fiber movement may also reduce the extent of clogging, or sludging. Like fouling, clogging or sludging of membrane channels results in reduced permeability and increased operating costs.

How fibers move when subjected to different sparging patterns (i.e. gas sparing intensity, intermittence of sparging, gas bubble size) and membrane configurations is not well understood. A potential tool in quantifying fiber movement inside a gas-sparged submerged hollow fiber membrane module is the use of a three-segment electrochemical shear probe. This probe allows for the determination of both the axial as well as the normal velocity relative to the membrane surface, as described by Sobolik et al. [177]. It may be possible to deduce information about fiber movement from the measured normal velocity. Once fiber movement can be quantified, the mechanism of fouling and clogging control as a result of this movement, as well as sparging and membrane configuration that affect fiber movement can be studied.

The impact of gas sparging on bulk fluid movement inside the membrane tank is also currently not well understood. When comparing the external side of the module compared to the internal side (i.e. between membrane cassettes), Nguyen et al. [128] reported a lower local vertical bubble velocity and local gas hold-up on the external side, using a bi-optical probe. The method of mounting the bi-optical probe inside the tank may possibly result in an interference of the flow field. The use of the electrochemical shear probe described in this thesis may be a suitable, non-invasive method in determining the bulk flow characteristics inside the membrane tank, and the required improvements in membrane configuration designs (i.e. cassette spacing, addition of baffles in tank etc.)

The experiments described in this thesis were performed using the electrolytes (Chapters 4 to 7) as well as bentonite water mixture (chapter 8), both of which are Newtonian in nature. The water matrix inside a membrane bioreactor for wastewater treatment can be non-Newtonian [13]. Now that the technique for shear measurements in a Newtonian system has been established in this thesis, the next step is to conduct shear measurements inside a non-Newtonian system by altering the viscosity of the electrolyte. This may possibly be accomplished by adding a polymeric substance to the ferricyanide and ferrocyanide solution, as suggested by Dumon et al. [185].

Relationship between Shear Stress and Fouling

In this thesis, interesting observations were obtained when investigating the effect of different types of transient shear stress on fouling rates, as described in Chapter 8. To be sure that the results obtained were real phenomena, and not as a result of apparatus artifact, a different shear apparatus should be designed and used to check if phenomena observed in Chapter 8 were indeed real.

To investigate the mechanisms of fouling control by the different types of transient shear profiles, direct observation methods can be used to confirm that the mechanisms of fouling control in high peak shear experiments was physical scouring, and that the fouling control mechanism in low peak and sustained peak shear is enhanced particle back-transport. Additionally, a better description of the relationship between transient shear stress and fouling is needed. As such, one needs to develop a way of properly describing the transient shear stress using a quantifiable parameter.

It will also be interesting to examine different effects of increased liquid flow in combination with transient shear stresss on fouling, i.e. sustained high shear stresses in combination with smaller variation in transient shear stresses, as shown in Figure 9-1. Finally, the effect of permeate suction on shear stress at membrane surface needs to be quantified (using flat sheet membrane).



Figure 9-1. Different types of transient shear profiles for future investigations

9.3 Engineering Significance

The main knowledge gap that leads to the present research study is that the hydrodynamic conditions inside a submerged hollow membrane module under gas-sparging are not well understood. As a result – the time-consuming and expensive process of pilot-testing is necessary before a full scale unit can be built. The research study conducted here narrows the above knowledge gap. The research study undertaken presents one of the most comprehensive investigations of the hydrodynamic conditions inside the submerged hollow fiber membrane module to date.

Based on these investigations – it is realized that the hydrodynamic conditions inside a submerged hollow fiber membrane are different than those of confined tubular membrane systems, which was initially hypothesized by some. These results provided significant insights regarding the interaction between bubbles and fibers, which yields knowledge regarding the relationship between gas sparing and fouling. It was also observed that different types of shear profiles exist inside the membrane module. These profiles are impacted by the membrane geometry as well as two-phase flow characteristics. Moreover, the different types of shear conditions result in different fouling, which suggests that different mechanisms are at play in controlling particle transport near the membrane surface. This information opens the opportunity for further investigation in terms of optimization of the gas –sparging system, or other shear-generating devices that create the shear conditions that offer the greatest benefit minimizing fouling, while minimizing the energy demand associated with generating these shear conditions.

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Appendices

Appendix A (Section 4.3.1)

Shear Probes Calibration – tubular pipe wall experiment

The measured shear signal (and hence calculated shear stress) are compared with theoretically calculated shear stress acting on a tubular pipe wall in single phase flow. Discrepancies between measured shear stress and theoretical shear stress are accounted for by applying a geometric calibration factor to the measured shear stress. The calculation of theoretical shear stress inside a tubular pipe is first presented. The used of the geometric calibration factor on measured shear stress is then described, and compared to the theoretically calculated shear stresses.

Calculation of Theoretical Shear Stress Acting on Pipe Wall

The following equations are used to calculate the shear stress acting on a pipe wall during single-phase laminar flow:

$$\tau = \frac{f_r \rho_L \mathbf{v}^2}{8} \tag{A1}$$

where f_r can be calculated as $\frac{64}{\text{Re}}$

Table A1 shows the theoretically calculated shear stress acting on the wall of a 9.9 mm (inner diameter) pipe (see Figure 4-4), for the range of liquid flow rates considered.

Liquid Flow Rate (l/min)	Velocity (m/s)	Re	f	Frictional headloss(m)	Theoretical Shear stress (Pa)
0.1	0.0217	218	0.2937	0.0007	0.018
0.2	0.0433	436	0.1469	0.0014	0.035
0.3	0.0649	654	0.0979	0.0021	0.053
0.4	0.0866	876	0.0734	0.0028	0.070
0.5	0.1083	1089	0.0587	0.00354	0.0875

Table A1 . Theoretical Single Phase Wall Shear Stress for a Pipe Diameter of 9.9 mm

Comparison between Measured Shear Stress and Theoretical Shear Stress without Geometric Correction Factor

In a flow cell which contains two probes (probe 1 and probe 2), the shear stress is measured individually for each probe. i.e. measure shear stress for flow rates 0.1 to 0.5 L/min using Probe1 only. Probe2 is not connected to the circuit. Conversely, when measuring shear stress using probe2, probe1 was not connected to the circuit. Table A2 shows the comparison between the measured shear stress and theoretically calculated shear stress. The measured shear stress was obtained using Equation 4-14 (note that Vcal in Equation 4-14 was the raw voltage signal, not the calibrated voltage signal).

Table A2. T	he comparison	between th	e measured	shear st	ress and	theoretically	calculated
shear stress.	No geometric	calibration	factor has l	been app	olied		

a) Probe 1							
							Difference
	Average						between Measured and
Flow Rate	Measured				Measured	Theoretical	Theoretical
(L/min)	Signal (V)	Ι	k	S	Shear (Pa)	Shear (Pa)	Shear Stress (%)
0.1	0.162	1.62E-06	2.44E-05	24.1	0.0241	0.0176	38
0.2	0.203	2.02E-06	3.06E-05	47.1	0.0471	0.0350	35
0.3	0.220	2.20E-06	3.31E-05	60.1	0.0601	0.0525	15
0.4	0.240	2.40E-06	3.62E-05	78.6	0.0786	0.070	12
0.5	0.259	2.58E-06	3.90E-05	97.9	0.0979	0.0875	12
b) Probe 2							
							Difference
							between
	Average						Measured and
Flow Rate	Measured				Measured	Theoretical	Theoretical
(L/min)	Signal (V)	Ι	k	S	Shear (Pa)	Shear (Pa)	Shear Stress (%)
0.1	0.167	1.66E-06	2.52E-05	26.4	0.026	0.017	51
0.2	0.202	2.02E-06	3.05E-05	46.8	0.046	0.035	34
0.3	0.225	2.24E-06	3.39E-05	64.4	0.064	0.0525	23

80.3

95.9

0.080

0.095

0.0700

0.0875

The difference between the theoretical and measured shear ranged from 10% to 51%. The biggest source of error in the experimentally measured shear is the geometry of the probe (see section 4.3.5). As seen in the microscope image of the probes (Figure 4-6),

3.65E-05

3.87E-05

0.4

0.5

0.243

0.257

2.41E-06

2.56E-06

15

10

the probes were not perfectly circular, (most likely ellipsoidal due to machining), and the surfaces of the probes were corrugated (seen as lines on probe surface) due to sanding. The total surface area available for the electrolytic reaction was greater than with a flat smooth circular probe with a diameter of 0.5 mm. A geometric correction factor was therefore applied to the diameter of the probes to minimize the difference between the theoretical and experimental shear stress.

Application of the Geometric Correction Factor

The geometric correction factor was applied to the measured probe diameter using the following relationship:

$$d_{cor} = d \times g_{cor}$$
 A2

Shear stress for flow rates ranging from 0.1 to 0.5 L/min were calculated using a range of g_{cor} values. The shear stresses were compared to the theoretical shear stress. The selection of the g_{cor} value was based on the g_{cor} which resulted in the *minimum* difference between the measured shear stress and the theoretical shear stress, as shown in Figure A1 and Figure A2.



Figure A1. Determination of g_{cor} based on the minimization of the difference between the measured and theoretical shear stress for probe 1



Figure A2. Determination of g_{cor} based on the minimization of the difference between the measured and theoretical shear stress for probe 2

Table A3 shows the comparison between the measured and the theoretical shear stress after applying $g_{cor} = 1.03$ for probe 1, and $g_{cor} = 1.06$ for probe 2.

Table A3.	The comparise	on between t	he measured	l and the	e theoretical	shear	stress	after
applying t	he geometric c	orrection fac	tor					

a) Probe 1, g _{cor}	= 1.03						
Flow Rate (L/min)	Corrected Signal (V _{cor})	Ι	k	S	Measured Shear (Pa)	Theoretical Shear (Pa)	% Difference
0.1	-0.162	1.620E-06	2.313E-05	21.038	0.021	0.018	20
0.2	-0.203	2.025E-06	2.891E-05	41.098	0.041	0.035	17
0.3	-0.220	2.196E-06	3.136E-05	52.412	0.052	0.053	0
0.4	-0.241	2.401E-06	3.428E-05	68.482	0.068	0.070	2
0.5	-0.259	2.583E-06	3.689E-05	85.316	0.085	0.088	3
b) Probe 2, g _{cor}	. = 1.06						
Flow Rate	Corrected Sign	nal				Theoretical	%
(L/min)	(V _{cor})	Ι	k	S	Measured Shear (Pa	a) Shear (Pa)	Difference
0.1	-0.167	1.669E-06	2.241E-05	19.73	3 0.020	0.018	13
0.2	-0.203	2.021E-06	2.714E-05	35.028	8 0.035	0.035	0
0.3	-0.225	2.247E-06	3.018E-05	48.16	9 0.048	0.053	8
0.4	-0.243	2.418E-06	3.247E-05	60.022	2 0.060	0.070	14
0.5	-0.257	2.565E-06	3.445E-05	71.67	5 0.072	0.088	18

Appendix B (Section 4.3.2)

Estimation of Test Fiber Probe Diameters

The diameters of Probe 1 and Probe 2, observed under the microscope and measured using the Motic Imaging software, were both 0.56 mm (radius 0.28 mm). These measured diameters were likely smaller than the true diameter, where curvurture exists. During mounting process, each probe was sanded down such that its surface is flush with the surface of the test-fiber. Curvature effect may be more prominent on the test fiber (diameter 1.7 mm) compared to the vertical pipe flow cell (diameter 9.9mm). Therefore, the "true" test-fiber probe diameter which takes into account the curvurture was calculated based on the measured diameter.

The calculation below shows approximately the true probe diameter, taking into consideration the curvature of the test fiber.

$$a^2 = b^2 + c^2 - 2bc \cos\theta \tag{B1}$$

$$s = r\theta$$
 (B2)

From Equation, B1, θ was calculated to be 0.67 rad. From Equation B2, was calculated to be **0.57 mm (radius 0.285 mm)**.



Figure B1. Test-fiber probe diameter

a) Top view of test fiber (test fiber radius r = b = c = 0.85 mm; a = platinum wire from diameter = 0.56 mm). b) Top view of the probe on the test fiber, dashed line is the circumference of the probe observed under the microscope, solid line is the true probe circumference, taking into consideration curvature effects.

Appendix C (Section 4.3.4)

Procedure for Measuring Shear Signal

The following is the procedure used for shear measurement of single and two phase flow in tubular pipes, described in Chapter 4:

- Purge distilled water with nitrogen gas for 15 minutes.
- Prepare the electrolyte solution. Pour solution into the gas/liquid separator tank and purge with nitrogen gas for 15 minutes.
- Place separator tank into water bath and wait for temperature to equalize to 20 °C.
- Rinse shear probes in flow cell or on fiber using distilled water, gently rub probe surface for 10 seconds with Q-tip and distilled water again.
- Clean anode (stainless steel pipe fitting) using a brush, clean with Q-tip and rinse with distilled water.
- Set up the flow cell and pipes (see Figure 4-4 and 4-9).
- Turn on the pump (flow rate 0.2 L/min) and let the electrolyte circulate in the loop, mixing with N₂ gas to purge system of oxygen for 15 minutes.
- Connect anode and probes to power supply.
- Set up is now ready for experimentation at desired liquid and gas flow rates.
- Use the Labview software for data acquisition
- Perform experiments behind the metal wire mesh, to limit interference from external electromagnetic frequencies.
- After each measurement, empty electrolyte in the pipe, remove flow cell, and repeat probe cleaning procedure.
- At the end of all experiments, drain the electrolyte from the apparatus and store in waste container.
- Rinse the apparatus with distilled water (by filling the air/liquid separator with water and then turning the pump on, circulating the fluid, then emptying the air/liquid separator and reversing the pump to withdraw any further liquid).

Appendix D (Section 4.3.4)

Instrument Bias and Signal Noise

The data acquisition box consisted of 2 channels available for acquiring shear signals. Each channel had an intrinsic instrument bias, where a small signal was measured even when the box was isolated from the electric circuit. The instrument bias for channels 1 and 2 are shown in Figure D1. The average instrument biases for channels 1 and 2 are 0.00979 and 0.0257 V, respectively. Measured shear signals from all experiments were corrected for these instrument biases (i.e. bias was subtracted from signal).



Figure D1. Instrument bias signals of Channel 1 and Channels 2. Signals measured when probes were not plugged into circuit

The signal noise for both channels 1 and 2 (i.e. variability in signal) in Figure D1 was 0.0018 V and 0.0015 V respectively. When measuring shear stresses during single phase liquid flow in the tubular pipe (up to Re=600) the variability in the signals ranged from 0.002 V (Re = 71) to 0.004 V (Re=600). The higher variability seen at higher flow rate

was likely due to liquid flow pulsing as a result of the flow pump. The noise level for all experiments was taken to be 0.002 V.

Appendix E (Section 4.3.7)

Limiting Current Test

An experiment was performed to check the limiting current of the electrochemical system. The experiments were conducted using the pipewall experiment set-up (see Figure 4-4), with varying liquid flow rates. The current through the circuit was measured for different applied potentials (starting at 0 mV and at increments of 50 mV to 400 mV). The time to reach equilibrium when changing the applied potential from 50 mV to 400 mV was found to be less than 2 seconds. Therefore, for each experiment, when the applied potential was increased or decreased by 50mV, 10 seconds passed before the data are collected to allow for adequate equilibriation time. The limiting current or diffusion condition was achieved when applying a potential in the range of 200mV to 400mV, as shown in Figure E1 for different Reynolds number.



Figure E1. The limiting current test at different Reynold's number. Experiment conducted using 0.003M ferricyanide, 0.006M ferrocyanide and 0.3M potassium chloride

Appendix F (Section 4.3.8)

Re-use of Electrolytes

Possible photochemical decomposition and oxidation reaction of ferricyanide and ferrocyanide may degrade the quality of the electrolyte. An experiment was conducted in three different days using the same electrolyte to check if the possible degradation of the electrolyte had an effect on shear measurements. Careful cleaning of the shear probe and the stainless steel pipe fitting was performed prior to each experiment. Figure F1 shows the three different experiments at different flow rates, conducted at 3 consecutive days using the same electrolyte. Although there were some differences between the measured signals for each different days, however, the differences between the measured signals of days 1 and 2. The differences between each signal were therefore likely due to a different controllable flow rate used each time, and not due to degradation of the electrolyte. This also shows that with careful cleaning of the probes and anode before each experiment, possible effects of electrode poisoning due to photochemical decomposition and oxidation reaction can be avoided, and the reaction byproducts in the electrolyte solution do not have a substantial effect on shear measurements.



Figure F1. Measurements conducted at Days 1, 2 and 3 to check for possible effects of electrolyte degradation

Appendix G (Section 4.4.1)

Indication of Flow Direction Using the Shear Probe

This section examines whether the measurements from probes in the pipe wall can indicated the direction of flow. Initially upward single phase flow in the vertical pipe (0.1 L/min and 0.2 L/min) were measured using probe 1 and probe 2. The measured, corrected signal is shown in Figure G1. The signal from probe 1 was higher than probe 2, which indicates direction of flow is from probe 1 to probe 2.

The direction of flow was then revered (downward flow) such that the flow is from probe 2 to probe 1. The measured signals from the probes are shown in Figure G2. The signal from probe 2 was now higher than probe 1 indicating direction of flow was from probe 2 to probe 1.



Figure G1. Measured, corrected shear signals of probe 1 and probe 2. The direction of flow is from probe 1 to probe 2



Figure G2. Measured shear signals of probe 1 and probe 2. The direction of flow is from probe 2 to probe 1

Increasing the flow rate was found to increase in the difference between the measurements of the probe 1 and probe 2, as shown in Figure G3. This observation can be attributed to the fact that increasing flow rate results in increasing the diffusional wake downstream of the first probe, thus provides more shielding of the second probe and thus the amount of species transferred to the surface of the second probe and reducing its measured current. As such, the differences in the signals observed during the passage of the gas slugs may also provide an indication of increased velocity near the pipe wall as liquid flow direction is reversed in the falling film region.


Figure G3. The difference in shear signals between probe 1 and probe 2 at different flow conditions

Appendix H (Section 6.3.1)

Surface Shear Profiles

The surface shear profiles at different radial sections at gas flow rates of 5 and 35 mL/min, for 1 mm and 2 mm nozzles, are shown in Figure H1.



Figure H1. Bubble-induced surface shear profiles at different radial sections of the fiber surfaces for gas flow rate of 5 mL/min, delivered through gas nozzle size of 1 mm

a) $\theta = 0$ degrees, b) $\theta = 45$ degrees, c) $\theta = 90$ degrees, d) $\theta = 135$ degrees, e) $\theta = 180$ degrees.



Figure H2. Bubble-induced shear profiles at different radial sections of the fiber surfaces, for a gas flow rate of 5 mL/min, delivered through gas nozzle size of 2 mm

a) $\theta = 0$ degrees, b) $\theta = 45$ degrees, c) $\theta = 90$ degrees, d) $\theta = 135$ degrees, e) $\theta = 180$ degrees



Figure H3. Bubble-induced shear profiles at different radial sections of the fiber surfaces, for gas flow rate of 35 mL/min, delivered through gas nozzle size of 2 mm

a) $\theta = 0$ degrees, b) $\theta = 45$ degrees, c) $\theta = 90$ degrees, d) $\theta = 180$ degrees

Appendix I (Section 7.2.1)

Normalization of Probe Diameter for P1-P7

In the experiments in Chapter 7 six different shear probes were used to measure simultaneously the shear forces within the fiber bundle. 40 shear probes were made, each shear probe had a slight different surface area for the other. When calibrating the shear probes, it was found that the shear signals obtained varied considerably from one probe to the next, for a given hydrodynamic condition. Figure I1 shows the variability of 15 randomly chosen probes. Of the 15 probes shown in Figure C1, six probes whose shear signals were closest to each other were chosen (probes B, H, N, L, A, F and D), as shown in Figure I2. The standard deviation of the average shear signals for these six probes was 0.009 V. A further calibration step was taken to standardize the diameter of all six probes.



Figure H1. The average shear signal of 15 randomly chosen probes at different flow rates. Large variability in shear signal was observed between the probes



Figure H2. Six probes whose shear signals were closest to each other chosen (probes B, H, N, L, A, F and D)

The signals of P2 to P7 were normalized against the signal of P1 using a normalization factor, where

Normalization factor =
$$\frac{I_{px}}{I_{p_1}}$$

Table I1 shows the calculated and normalized diameter of the probes. The measured signals of each probe were also corrected for instrument bias. Table I2 shows the instrument bias for each channel of the data acquisition box, and the corresponding instrument bias.

FLOW (L/min)	H (P1)	D (P2)	L (P3)	F (P4)	N (P5)	B (P6)	A (P7)
0.1	0.5	0.532	0.490	0.485	0.502	0.498	0.507
0.2	0.5	0.522	0.489	0.483	0.499	0.501	0.500
0.3	0.5	0.518	0.492	0.482	0.499	0.505	0.494
0.4	0.5	0.510	0.490	0.477	0.495	0.503	0.483
0.5	0.5	0.510	0.505	0.482	0.496	0.504	0.483
Average Diameter	0.5	0.518	0.493	0.482	0.499	0.502	0.493
Normalized	1	0 021	1 000	1 077	1 006	0 001	1 000
Diameter	1	0.931	1.028	1.077	1.000	0.991	1.020

Table I1. Calcualted and normalized probe diameter (mm)

Table I2. Instrument Bias for Channels 1 to 7

Channel	Probe	Instrument Bias (V)
1	1	0.0095
2	2	0.0334
3	3	0.00894
4	4	0.0056
5	5	0.00834
6	6	0.01452
7	7	0.00029

Appendix J (Section 7.3.1)

Total Peak Count

The total peak count of the P1 to P7 when sparging through the 3 mm nozzle, is shown in Figure J1.



Figure J1. Total peak count of P1 to P7 during sparging at different gas sparging rates through 3 mm nozzle

(a) Nozzle A, (b) nozzle B, (c) nozzle D, (d) nozzle E

Appendix K (Section 8.2.1)

Leveling of the Shear Apparatus

The leveling of the shear apparatus was found to be very important in creating a consistent shear profile at different radial positions of the tank. Initially, shear signals were measured using the 1 blade block impeller at radial positions I, II, III and IV, seen in Figure 8-3. The shear apparatus was not checked to see if the structure was leveled with the surface of the lab bench. The measured shear signals of the unleveled apparatus are shown in Figure K1. With the passage of the blade, shear signal were higher than the baseline shear signal (approximately 0.11 V). Shear signal at position I was also observed to be higher than shear signals at position I. This suggested that either the entire apparatus, or the impeller was not leveled relative to the surface of the bench. Careful checking of the apparatus showed that the motor which was connected to the impeller shaft was not leveled (one corner was higher elevated by approximately 0.8 mm). When the shaft was carefully re-leveled, the shear signals at different radial positions were more consistent, as shown in Figure K2.



Figure K1. Shear stresses at radial positions I, II, III and IV using the 1 blade block impeller. Shear apparatus was not levelled with lab bench



Figure K2. Shear stresses at radial positions I, II, III and IV using the 1 blade block impeller. Shear apparatus was leveled with lab bench

Appendix L (Section 8.2.1)

Pressure Transducer Calibration

Calibration curve of the pressure transducer is shown in Figure L.



Figure L. Pressure Transducer Calibration Using Portable Calibrator (DPI 601, Druck Inc.)

Appendix M (Section 8.2.1)

Bentonite Particle Size Distribution

Bentonite particle size distribution of the water matrix, measured using the Mastersizer Hydro 2000S (Malvern), is shown in Figure M. Mastersizer's built-in sonicator used to disperse particles prior to analysis.



Figure M. Bentonite particle size distribution (concentration 0.5 g/L) using the Mastersizer Hydro 2000S (Malvern)

Appendix N (Section 8.2.1)

Clean Water Experiments

Initially, clean water experiments were performed every time after each cleaning cycle. The order of the experiments performed is shown in Table N. Figure N shows the results of 5 clean water experiments after 5 different filtration experiments. The clean water filtration pressure between 10 minutes and 30 minutes are shown. Filtration pressures prior from 0 minutes to 10 minutes are not shown because during that period of time the pressure is increasing to the equilibrium pressure. It can be seen that the filtration pressure for all five experiments were between 3 and 3.3 psi. Performing the clean water filtration experiments after each successive filtration experiments did not result in an increased clean water pressure (i.e. after experiment "50-low-40-4-A2", clean water pressure was 3.2 psi (experiment order 2); after experiment "50-low-50-1-A1" (experiment order 4), clean water pressure was 3.0 psi. This suggests that the cleaning procedure was effective in removing cake foulants from the membrane surface. The variation in clean water pressures were therefore considered due to various other factors during filtration (i.e. variation in permeate flow). Because the clean membrane permeability was relatively constant, for the subsequent experiments, the clean water filtration experiments were performed after every 5th or 6th filtration experiment to check that the clean membrane permeability had been recovered.

Experiment Order	Filtration Experiment Name	Clean Water Experiment Name
1	50-block-40-4-A2	50-block-40-4-A2-clean
2	50-low-50-4-A2	50-low-50-4-A2-clean
3	50-low-50-2-A3	50-low-50-2-A3-clean
4	50-low-50-1-A1	50-low-50-1-A1-clean
5	50-low-50-1-A2	50-low-50-1-A2-clean

Table N. Order of filtration experiments after which clean water experiments were performed



Figure N. The results of 5 clean water experiments after 5 sequential filtration experiments

Appendix O (Section 8.3.1)

Occurrence of Flow Reversal at Membrane Surface Using Block Shear Blades

A set of experiments, using the double-probe was performed to check whether flow reversal occurred near the membrane during the passage of the block shear blades. The shear measurements are shown in three replicates in Figures Oa,b and c. Two shear curves are shown on each figure – P1 and P2. P1 was the shear probe placed upstream of the fluid flow, and P2 was the shear probe placed downstream of the fluid. If the signal of P1 is greater than the signal of P2, then no flow reversal occurred. Conversely, if the signal of P2 is greater than the signal of P1, flow reversal occurred near the membrane surface. All of the figures below show that no flow reversal occurred near the membrane surface.





Figure O. Shear measurements using double shear probe during the passage of the block shear blades, in three replicates (a, b and c). P1 is the shear probe placed upstream of the direction of flow. P2 is the shear probe placed downstream of the direction of flow

Appendix P (Section 8.3.4)

Linear Regression of Pressure Curves

The fouling rates used for the correlation analysis in Chapter 8 were estimated using linear regression of the pressure curves of all experiments conducted. Below are figures the linear regressions of each experiment. The slope of the line was used as the fouling rate for each experiment.

a. The 0.2 g/L Experiments

1. Low Peak Shear Experiments



Figure Pa1. Linear regression of the pressure curves of the 0.2 g/L low peak shear experiments

4 blade low peak experiments – Low 4a and Low 4b. 2 blade low peak experiments – Low 2a and Low 2b. 1 blade low peak – Low 1a

2. High Peak Shear Experiments



Figure Pa2. Linear regression of the pressure curves of the 0.2 g/L high peak shear experiments

4 blade high peak experiments – High 4a and High 4b. 2 blade high peak experiments – High 2a and High 2b. 1 blade high peak - High 1a and High 1b

3. Block Shear Experiments



(Figure title follows next page)



Figure Qa3. Linear regression of the pressure curves of the 0.2 g/L block shear experiments

4 blade block experiments Block 4a, Block 4b and Block 4c. 3 blade block experiments Block 3a, Block 3b and Block 3c. 2 blade block experiments Block 2a, Block 2b and Block 2c. 1 blade block experiments Block 1a and Block 1b

4. Single Phase Shear Experiments



Figure Pa4. Linear regression of the pressure curves of the 0.2 g/L single phase shear experiments

Single a, Single b and Single c

b. The 0.5 g/L Experiments

1. Low Peak Shear Experiments



Figure Pb1. Linear regression of the pressure curves of the 0.5 g/L low peak shear experiments

4 blade low peak experiments – Low 4a, Low 4b and Low 4c. 2 blade low peak experiments – Low 2a and Low 2b. 1 blade low peak – Low 1a and Low lb

2. High Peak Shear Experiments



Figure Pb2. Linear regression of the pressure curves of the 0.5 g/L high peak shear experiments

4 blade high peak experiments – High 4b and High 4c. 2 blade high peak experiments – High 2a and High 2b. 1 blade high peak - High 1a and High 1b

3. Block Shear Experiments



Figure Pb3. Linear regression of the pressure curves of the 0.5 g/L block shear experiments

4 blade block experiments Block 4b and Block 4c. 3 blade block experiments Block 3a, Block 3e and Block 3f. 2 blade block experiments Block 2b and Block 2c. No 1 blade block experiments are available

4. Single Phase Shear Experiments





Single a, Single b and Single c

Appendix Q (Section 8.3.4)

The fouling rates obtained from the regression analyses in Appendix P were plotted against the different combinations of shear parameters, as seen in Figure Q1.

Figure Q1. The calculated pressure increase rate plotted against the different shear parameters (continued next page)



