MEMBRANE FOULING IN INDUSTRIAL ANAEROBIC MEMBRANE BIOREACTORS

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Abstract

Anaerobic membrane bioreactors (AnMBRs) offer a suitable alternative to existing lagoon and high-rate granular treatment options for high solids and fatty industrial wastewaters. AnMBR fouling is however, a major limitation to industrial applications, particularly in the treatment of complex, particulate substrate. Model analyses that integrate the multiple controlling factors of fouling offer a better understanding of controlling mechanisms than experimental analysis, however, existing membrane bioreactor (MBR) models are limited to simple hydraulic profiles and do not take into account complex reactor hydrodynamics. In response, this thesis investigated the effect of particulate fouling on AnMBR performance and identified the impact of reactor hydrodynamics on membrane fouling for the development of a holistic MBR optimisation model. Mechanistic fouling analysis identified that protein hydrophobicity and compressibility were key contributors to its high fouling propensity. Protein also showed high reactivity with lipids causing synergistic fouling behaviour in protein-lipid mixtures and a surfactant effect on lipids, changing the fouling mechanism from irreversible pore fouling in pure lipids to cake fouling in protein-lipid mixtures. CFD modelled shear was integrated into a distributed parameter model to simulate membrane fouling profile and flux distribution. Incorporation of a flux-pressure feedback loop allowed for the simulation of flux-step experiments and the prediction of critical flux. This model demonstrated a wide variety of applications including, for the identification of complex hollow fibre shear and the effect of operating conditions, such as total solids, sparge rate and filtration strategy, which can be directly applied to MBR design, optimisation and cost analyses.
Declaration by author

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This paper has been modified and incorporated in this thesis as Chapter 4.

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<tr>
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Contributions by others to the thesis

I would like to acknowledge the contribution of Shao Dong Yap who conducted pilot-scale anaerobic membrane bioreactor flux-step analyses discussed in Chapters 4 and 5.

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LIST OF ABBREVIATIONS

Acronyms
AnMBR   Anaerobic membrane bioreactor
CFD    Computational fluid dynamics
COD    Chemical oxygen demand
EPS    Extracellular polymeric substances
FOG    Fats, oils and grease
FS    Flat sheet
HF    Hollow fibre
HRAT    High rate anaerobic treatment
LMH    Litres per square metre per hour
MBR    Membrane bioreactor
MLSS    Mixed liquor suspended solids
PDE    Partial differential equation
PSD    Particle size distribution
SMBR    Submerged membrane bioreactor
SMP    Soluble microbial products
TMP    Transmembrane pressure
TS    Total solids
UASB    Upflow anaerobic sludge blanket
WW    Wastewater

Nomenclature

\[ \alpha \]  
stickiness coefficient [ ]

\[ \beta \]  
erosion rate coefficient of cake [ ]

\[ \gamma \]  
shear strain [s\(^{-1}\)]

\[ \gamma' \]  
cake layer compression coefficient [kg/(m\(^3\)s)]

\[ \varepsilon \]  
dissipation rate of kinetic energy per unit mass

\[ \theta_f \]  
filtration period in a cycle [min]

\[ \kappa \]  
von Karman’s constant \( \approx 0.4 \) for smooth walls

\[ \mu \]  
dynamic viscosity [Pas]

\[ \mu_{eff} \]  
effective viscosity [Pa.s]

\[ \mu_t \]  
turbulence viscosity [Pa.s]
\( \mu_{t,G} \) dynamic turbulent viscosity of the gas phase [Pa.s]
\( \mu_{t,L} \) dynamic turbulent viscosity of the liquid phase [Pa.s]
\( \mu_{t,P} \) particle-induced turbulent viscosity [Pa.s]
\( \mu_{t,S} \) shear-induced turbulent viscosity [Pa.s]
\( \rho \) density [kg/m\(^3\)]
\( \sigma \) surface tension coefficient [N/m]
\( \tau \) shear stress [Pa]
\( \tau_W \) wall shear stress [Pa]
\( \tau_Y \) yield stress [Pa]
\( \nu \) kinematic viscosity [m\(^2\)/s]
\( \nu_{t,G} \) kinematic turbulent viscosity of the gas phase [m\(^2\)/s]
\( \nu_{t,L} \) kinematic turbulent viscosity of the liquid phase [m\(^2\)/s]
\( \phi \) general scalar or vector quantity per unit volume
\( \varphi \) general scalar or vector quantity
\( \bar{\varphi} \) mean component of scalar quantity
\( \bar{\varphi}_\alpha \) Favre-averaged scalar quantity
\( \bar{\varphi}_\alpha \) mean (time averaged) component of the scalar quantity
\( \varphi' \) fluctuating component of scalar quantity
\( \chi \) diffusivity of fluid at rest [m\(^2\)/s]
\( \omega \) fibre shear enhancement factor [ ]
\( C_{\mu}, C_{\varepsilon 1}, C_{\varepsilon 2}, \sigma_k \text{ and } \sigma_\varepsilon \) empirical k-\( \varepsilon \) turbulence coefficients
\( C_{\alpha\beta} \) momentum transfer coefficient
\( C_D \) drag coefficient [ ]
\( C_L \) lift coefficient [ ]
\( d_B \) bubble diameter [mm]
\( d_p \) particle diameter [\( \mu \)m]
\( D_F \) drag force [N]
\( E \) additive constant \( \approx 9.8 \) for smooth walls
\( E_0 \) Eotvos number [ ]
\( F \) momentum flux
\( F_D \) diffusive flux
\( F_C \) convective flux
\( F_{TD} \) turbulent dispersion force [N]
\( J \) filtration flux [LMH]
K     viscosity consistency [Pa.S]
Ki     lift coefficient [m]
k      turbulence kinetic energy
LF     lift force [N]
M     Morton number [ ]
\(M_\alpha\)  force acting on phase \(\alpha\) due to interaction with other phases [N]
\(M_c\)  accumulated cake mass [g/m²]
p or P  pressure [Pa]
P'     transmembrane pressure at which \(r_{c0}\) doubles [Pa]
P_k  turbulence production due to viscous forces
Pr  turbulent Prandtl number [ ]
Qv  volume source
QS  surface source
\(r_\alpha\)  volume fraction of phase \(\alpha\)
\(\bar{r}_\alpha\)  mean (time averaged) volume-fraction of phase \(\alpha\)
Re  Reynolds number [ ]
Re_B  bubble Reynolds number [ ]
R  total resistance [m⁻¹]
\(R_c\)  cake resistance [m⁻¹]
\(R_m\)  membrane resistance [m⁻¹]
\(r_c\)  specific cake resistance [m/kg]
\(r_{c0}\)  initial specific cake resistance [m/kg]
SM  momentum source
\(S_{MS_\alpha}\)  mass source
T  temperature [°C]
u, v, w  velocity components in x, y, z directions
\(u_\tau\)  skin friction velocity [m/s]
\(V_f\)  volume of filtrate [m³]
\(V_G\)  gas flow rate [m³/s]
y*  non-dimensional distance from the wall [ ]
Anaerobic digestion has been successfully implemented in the treatment of sludge from municipal treatment as well as in high organic strength industrial wastewaters. The main advantages of anaerobic processes is that no external oxygen is required for microbial growth, and the process produces energy-rich biogas. Agro-industrial wastewater, for example from food and animal processing industries, is high in organic content, making it ideal for anaerobic treatment processes aimed at energy recovery [1]. Anaerobic lagoons are a default treatment option for the treatment of agro-industrial wastewater; however these have major disadvantages; including variable biogas production, large footprints (and hence poor volumetric loading), poor biogas capture and high desludging costs. Engineered reactor systems are easier to control and offer a higher rate of anaerobic treatment and hence greater volumetric loading than lagoons. A reactor's ability to effect high rate anaerobic treatment (HRAT) is dependent on sufficient retention of the slow growing anaerobic biomass, which is achieved in HRAT technologies by sludge granulation, for example, in upflow anaerobic sludge blanket (UASB) reactors [2]. However, high solids and lipid concentrations typically found in food and animal processing wastewater, can impede granulation [2, 3] and lead to decreased settleability and hence lower biomass (and particulate substrate) retention.

Membrane bioreactors (MBRs) replace gravity settling with membrane filtration and are therefore not dependent on sludge settleability for biomass retention. MBRs are classically aerobic; i.e. they integrate membrane filtration with the activated sludge process, and have been widely applied in domestic and industrial wastewater treatment [4-6]. Anaerobic MBRs (AnMBRs), are a more recent development [4]; that combine membrane filtration with anaerobic digestion, hence introducing the advantages of anaerobic digestion, i.e. biogas production and elimination of oxygen, into MBR technology. The combination of anaerobic digestion for energy recovery and membrane filtration for solids retention makes AnMBRs an attractive option for the treatment of high solids and lipids containing waste streams [7].

1.1 MBR FUNDAMENTALS

1.1.1 MBR configuration

MBRs can be configured in either a sidestream (crossflow membrane) or a submerged design. In a sidestream MBR (Figure 1-1a), mixed liquor from the treatment vessel is pumped to an externally
housed membrane which separates suspended solids (retentate) from treated effluent (permeate) before returning the retentate to the main digester. In a submerged MBR (SMBR) (Figure 1-1b), a membrane filter is immersed within the main treatment vessel to retain suspended solids within the process. In both configurations, membrane fouling is controlled via shearing of the membrane surface, either by liquid crossflow in the sidestream configuration, or via gas sparging in the submerged configuration. In submerged AnMBRs, biogas produced from anaerobic treatment can be recirculated as sparger gas, minimising (or even eliminating) the requirement of external gas.

Figure 1-1: MBR configurations, including (a) sidestream membrane bioreactor and (b) submerged membrane bioreactor (SMBR).

1.1.2 Membrane filtration and fouling kinetics

Membrane filtration rate or flux (permeate flowrate per unit membrane, L/m²-h or LMH), is directly proportional to transmembrane pressure (TMP) and is related by liquid viscosity and total resistance via Darcy’s Law (Equation 1-1).

\[
\text{TMP} = \text{Flux} \times \text{viscosity} \times \text{Total resistance}
\]  

(1-1)

Total resistance is controlled by the permeability of the membrane as well as of the fouling layer that forms on the surface. Resistance-in-series is most commonly assumed, where total resistance is the sum of the membrane and fouling layer resistance. Membrane fouling is strictly defined as the build-up of generally insoluble material on or within the membrane [8]; and therefore also implies a loss in permeability (increase in total resistance).
Fouling rate is governed by the balance of flux-induced drag force and membrane shear force as illustrated in Figure 1-2. In steady-state operation (Figure 1-2a) flux-induced drag and shear are in-balance and the fouling layer does not grow over time, hence maintaining a constant total resistance. This leads to a stable flux-TMP relationship in accordance with Equation 1-1. Flux below which steady-state operation prevails is generally termed the ‘critical flux’ of the system.

An increase in flux promotes convective solids transport towards the membrane and accelerates solids deposition (Figure 1-2b) until either a new steady-state balance is reached or, if flux-induced drag greatly exceeds shear, the membrane is completely fouled and no further filtration is possible. In highly compressible filtration cakes, typically found in biological wastewater treatment, fouling layer resistance increases with applied pressure which can further accelerate membrane failure (Figure 1-2c).

Figure 1-2: a) Steady-state operation; b) accelerated cake deposition and c) compressible cake deposition (adapted from Yoon, 2012 [9])

High rate AnMBR treatment (high organic loading rate and short hydraulic retention times) requires a sufficiently large concentration of active in-reactor biomass (total solids) and large membrane flux. However, as stated before, large membrane fluxes can increase membrane fouling and destabilise long-term membrane operation. Membrane shear can be enhanced to counter the larger drag force induced by higher fluxes. However, this requires greater energy input in the form of liquid crossflow pumping or gas sparging, especially in concentrated sludge with high viscosity and density. Overall process flowrates can be enhanced by increasing membrane surface area, however this has associated capital costs (membranes are expensive and can comprise a significant proportion of overall capital costs [5, 6]). Therefore, the high rate treatment potential of an AnMBR and hence its viability as a treatment option for industrial applications, is ultimately affected by the
system’s membrane fouling propensity; with operating TS concentration and membrane flux (HRT) limited by the need to manage membrane fouling.

1.1.3 Factors affecting membrane fouling and AnMBR filtration performance

Membrane fouling is a widely studied phenomenon in MBR research (a SCOPUS search identified fouling as a keyword in 51% of ‘membrane bioreactor’ studies). A major driver for this research is the impact of membrane fouling on overall costs and plant productivity. Despite the heavy research focus on membrane fouling, there is still a lack of fundamental understanding and universal agreement on the causes of fouling [5]. The main factors that encourage fouling in aerobic MBRs have been identified as the biomass (mixed liquor total solids) concentration, particle size, sludge rheology and concentration of bound or suspended extracellular polymeric substances (EPS) [5, 10], but attempts to correlate these parameters with fouling have so far provided inconsistent results.

Very little fundamental research is conducted in anaerobic MBRs compared with aerobic MBRs [11, 12], and therefore, it is unknown whether findings from aerobic MBR studies are applicable to anaerobic MBRs, or whether there exist major differences in fouling mechanisms between the two configurations. Martin-Garcia et al [13], report that AnMBR fouling is more likely due to the presence of higher population of particulates in AnMBRs compared with aerobic MBRs. Food and animal processing waste streams especially contain high concentrations of particulate proteins, carbohydrates and lipids. These slowly degradable particulates can accumulate over long periods in AnMBR systems [14] and will therefore affect bulk filtration properties. Substrate composition also plays a key role in membrane fouling in AnMBRs. Proteins and lipids in particular have shown high fouling propensity in AnMBRs treating food and animal processing wastewater [15-17]. However; fundamental analyses that precisely identify the effect of particulate substrate and substrate composition on AnMBR fouling is lacking in literature.

Hydrodynamics also play a major role in MBR optimisation via membrane shear. Hydrodynamics within a reactor or membrane module is highly sensitive to its internal geometry. Therefore, empirical correlations between membrane fouling and operating conditions are design specific; and the correlations obtained in one design cannot be easily applied in vastly different designs or scales; due to the differences in hydrodynamics (and hence shear) between these systems. Drews (2010) [5] in a major membrane fouling review concluded that lab-scale studies are often of limited value to full-scale applications due to the non-representative hydrodynamics of the lab-scale configuration. Despite this, the effect of hydrodynamics is often overlooked in membrane fouling studies, most likely due to the difficulty in experimental characterisation of hydrodynamics.
The various factors affecting membrane fouling are also generally studied in isolation, as most experimental analyses are limited to studying the impact of a small number of process variables at a given time. However membrane fouling is a complex phenomenon and requires a multi-variable approach [18] that takes into account the combined effect of reactor design, operating conditions and wastewater characteristics to understand, predict and control (Figure 1-3). Model based analysis provides an alternative tool for fouling characterisation and effective fouling control, as the controlling factors can be integrated into an overall model framework which allows for determination of platform independent fouling characteristics and a better fundamental understanding of controlling mechanisms. Existing membrane fouling models are however limited to simple hydraulic profiles [19-25] and do not take into account complex reactor hydrodynamics. This is particularly important in submerged MBR configurations where the overall tank design can have a major impact on membrane shear.

![Diagram](image)

**Figure 1-3: Factors affecting membrane fouling and membrane filtration performance.**

The overall aim of this PhD project was to build an improved fundamental knowledge of hydrodynamics and fouling behaviour in agro-industrial (cattle slaughterhouse) AnMBRs.

This project aimed to address the identified gaps by:

(i) Characterising the hydrodynamics of a pilot-scale submerged AnMBR using computational fluid dynamics (CFD) to predict the impact of operational strategy on membrane shear

(ii) Identifying the fouling behaviour of both pure and complex slaughterhouse wastewater and the effect of particulate substrate composition on membrane fouling and;
Integrating the outcomes of (i) and (ii) to enable AnMBR optimisation that takes combined consideration of reactor hydrodynamics and membrane fouling kinetics.

1.2 RESEARCH OBJECTIVES AND APPROACH

Objective 1: AnMBR hydrodynamic characterisation (Chapter 2)

The first objective was to characterise reactor hydrodynamics, including membrane shear, in a pilot scale AnMBR. This was achieved by the use of multiphase computational fluid dynamics (CFD); based on its successful implementation by a number of researchers for predicting the effect of wastewater treatment process design features on the hydrodynamics of full scale installations [26-31].

Objective 2: Particulate fouling and effect of substrate composition (Chapter 3)

This objective investigated individual and combined fouling behaviour of three model substrate which are representative of the insoluble proteins, carbohydrates and lipids found in high solids and fatty cattle slaughterhouse wastewaters. These include egg white powder (protein), α-cellulose (carbohydrate) and animal fat (lipids). Fouling behaviour was identified by lab-scale filtration analyses in individual, binary and ternary combinations of protein, carbohydrate and lipids. Measured particle size and hydrophobicity and estimated cake properties (resistance and compressibility) provided supporting explanations for observed variations in behaviour.

Objective 3: SMBR fouling model development and model based analyses (Chapters 4 and 5)

CFD model shear estimated in objective 1 was integrated into the development of a multidimensional membrane fouling model to predict cake accumulation on a flat sheet submerged membrane reactor (Chapter 4). The model was used for further analyses into the identification of complex hollow fibre shear; and the effect of operating conditions (TS and gas sparge rate) and filtration strategy on membrane performance.
1.3 REFERENCES


2. COMPUTATIONAL FLUID DYNAMICS FOR MEMBRANE BIOREACTORS

ABSTRACT

Computational fluid dynamics (CFD) is widely used in many process based industries, including wastewater treatment, and is employed when experimental determination of flow behaviour is difficult, which is often the case in complex wastewater flows and treatment processes. The objective of CFD modelling in this thesis was to calculate membrane shear in a membrane bioreactor (MBR). The commercially available ANSYS CFX software was used which applies a finite-volume numerical method to solve fluid flow equations. Gas-liquid flows were solved using a two-fluid (Eulerian-Eulerian) approach, with each fluid described by its own set of governing equations and linked by interphase momentum transfer models. The CFD model approach included industry validated empirical submodels for turbulence (k-ε) and interphase momentum transfer (Schiller-Nauman and Grace drag, Tomiyama lift, Sato enhanced turbulent dispersion and Favre-averaged drag force models). Wastewater sludge in a two-phase approximation is treated as homogenous (i.e. no local variations in total solids) and can hence be modelled using a single rheological model, obtained from literature. For three-phase simulations, the third solids phase is modelled using the Algebraic Slip Method. Slip velocity for anaerobic MBR (AnMBR) sludge was measured in this chapter using laboratory zone settling rate tests. The CFD model was validated against a literature obtained experimental setup which measured bubble shear on a Teflon fibre submerged in a 16 L water tank. Two and three phase model assumptions were also compared; with minimal differences in velocity and shear profiles between the two models. The three-phase simulation took considerably longer to reach completion (82 hours) than the two-phase simulation (2 hours). Therefore, the two-phase simulation was considered appropriate for further CFD analyses of the pilot scale AnMBR. Further model applications include the application of CFD shear into a multidimensional membrane fouling prediction model.
2.1 INTRODUCTION

Computational fluid dynamics is the computer-based simulation of fluid flow, heat and related transport phenomena in a system and is widely applied in a range of industries where knowledge of fluid flow (hydrodynamics) is important. In wastewater (WW) treatment processes, hydrodynamics inside a reactor greatly affects treatment capability [1]. Lab-scale WW performance is often different from full-scale performance due to non-representative hydrodynamics at the two scales [2]. In complex reactor configurations, such as the submerged membrane bioreactor (SMBR), the various processes for membrane filtration, membrane fouling, biokinetics and reactor hydrodynamics are interlinked, and optimal SMBR performance requires combined consideration of all these processes [3].

Hydrodynamics is often overlooked in WW studies [4] as it is difficult to determine in practice. Monitoring techniques that exist for hydrodynamic determination are difficult to implement in large scale wastewater treatment systems due to transient or varying flows and opaque fluids. Measurement techniques are also restricted by the lack of space in reactor systems [5]. Modelling of hydraulics is also complex. The transport equations of fluid, heat and mass transfer are complex, second-order partial differential equations which often have non-linear components [6]. The field of fluid dynamics is mathematically intensive, involving partial differential equation theory, vector and tensor analysis and non-linear methods and has both benefitted from and contributed to the development of finite-difference, finite-element and finite-volume numerical techniques [7]. The ability to numerically solve these has increased with advancing computer technology [8]. Hence, recent advances in computer hardware have enabled practical use of CFD in wastewater modelling, and a number of researchers have successfully utilised and implemented CFD as a tool for predicting the effect of wastewater treatment process design features on the hydrodynamics of full scale installations [4, 9-13].

This PhD project uses CFD as a modelling tool, in particular for the estimation of membrane shear, which is essential for the prediction of membrane fouling kinetics. This chapter introduces key concepts in CFD modelling, and specifics of the CFD submodels used in this study. A large number of empirical sub-models are available to the CFD modeller. In this thesis, sub-models were chosen based on how widely validated they are in literature and whether they are applicable to wastewater flow in an anaerobic membrane bioreactor (AnMBR); i.e. whether they were developed in conditions similar to those in an AnMBR that typically comprises incompressible fluid, bubbly flow, multiple phases, anaerobic sludge etc. The model is applied and validated against shear
measurement experiments conducted by Bèrubè et al., (2006) [14]. Finally, two and three-phase CFD model behaviour was tested and compared on the pilot scale AnMBR.

2.2 CFD FUNDAMENTALS

2.2.1 Modelling strategy

The CFD approach used in this thesis is mesh based and uses an Eulerian frame of reference. Wastewater generally comprises multiple phases: dispersed solid particles and gas bubbles in a liquid wastewater continuum. CFD studies of WW problems generally do not model the solids phase, and instead approximate the reactor fluid as a two-phase (gas-liquid) mixture [4, 11, 12, 15, 16]. The liquid phase is linked to total solids concentrations via its viscosity. Brannock et al (2010) [12] suggest that a two fluid approximation for most wastewater fluid flow problems is appropriate as the positive buoyancy experienced by bubbles is much greater than the negative buoyancy experienced by settling sludge. Liu et al (2015) [17], also found that the relaxation times for solid particles to reach liquid velocity is negligible. This assumption greatly simplifies the fluid-flow problem as only two sets of conservation equations and related interphase transfer equations are to be solved. Le Moullec (2010) [16] however highlight the usefulness of considering all three phases in hydrodynamic determination as this will enable visualisation of the settling profile of the solids phase, mixing efficiency and mass transfer aspects.

In this chapter, reactor fluid was approximated as a two-fluid (Eulerian-Eulerian) mixture of dispersed gas bubbles in a liquid continuum. Therefore each fluid is described by its own set of conservation equations (mass, momentum and turbulence) coupled by momentum transfer equations (mass transfer was not considered). Two-phase results are also compared with a three-phase model (two-fluid with an algebraic slip model to separate solids and liquids).

2.2.2 Governing equations of flow

Fluid mechanics equations are set in the form of a general transport equation which describes the conservation of a scalar per unit volume, \( \phi \) (or vector property, \( \vec{\phi} \)) in an arbitrary control volume. The differential conservation form of the general transport equation is shown in Equation 2-1. This equation states that the rate of change of \( \phi \) inside an arbitrary control volume is equal to the boundary fluxes in \( \phi \) (\( \vec{F} \)), plus volume (\( Q_v \)) and boundary (\( Q_s \)) source terms.

\[
\frac{\partial \phi}{\partial t} = -\nabla \cdot \vec{F} + Q_v + \nabla \cdot \vec{Q}_s
\]  

(2-1)
Flux, $\mathbf{F}$, has diffusive ($F_D$) and convective ($F_C$) components. $F_D$ is expressed by Fick’s law (Equation 2-2); where $\chi$ is the diffusivity of the property in fluid at rest [m$^2$/s]. $F_C$ is the quantity of $\varnothing$ transported in a flow with velocity, $v$ [m/s] (Equation 2-3).

$$F_D = -\chi \nabla \varnothing$$

$$F_C = v \varnothing$$

Substitute the above into Equation 2-1 to obtain the general transport equation in Equation 2-4.

$$\frac{\partial \varnothing}{\partial t} + \nabla \cdot (v \varnothing) = \nabla \cdot (\chi \nabla \varnothing) + Q_v + \nabla \cdot Q_s$$

[Unsteady term + Advection = Diffusion + Source]

Let $\varnothing = \rho \varphi$, where $\rho$= specific mass of the fluid. By setting $\varphi$ equal to 1, the mass transport equation is obtained. By setting $\varphi$ equal to $v$ and setting appropriate diffusion coefficients, the momentum transport equations can be obtained.

The compressible form of mass conservation or continuity equation is detailed in Equation 2-5, and therefore expresses that mass is transported through convection only (i.e. no diffusive fluxes exist for mass transport, and any sources/sinks due to chemical reactions are ignored).

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho v) = 0$$

Liquids are generally incompressible, i.e., their density is unchanged. Therefore, for the purposes of wastewater CFD modelling, an incompressible continuity equation can be adopted (Equation 2-6).

$$\nabla \cdot \mathbf{v} = 0$$

By expanding the velocity vector, $\mathbf{v}$ into its x, y and z-direction components ($u$, $v$ and $w$, respectively), Equation 2-6 can be expressed as the incompressible continuity equation in Cartesian coordinates (Equation 2-7);

$$\frac{\partial u}{\partial t} + \frac{\partial v}{\partial x} + \frac{\partial w}{\partial z} = 0$$

The momentum equation in x, y and z-directions are similarly obtained and are expressed in Equation 2-8.

$$\frac{\partial (\rho v)}{\partial t} + \nabla \cdot (\rho v \otimes \mathbf{v} + p \mathbf{I} - \mathbf{t}) = S_M$$

Where; $p \mathbf{I}$= isotropic pressure tensor and $\mathbf{t}$= viscous shear stress tensor.
[Note: $\otimes$ = the tensor product of two vectors (also known as a dyadic when both vectors have the same dimension)]

The *incompressible momentum equation in Cartesian coordinates* in the x-direction (after expanding the relevant terms), is given in Equation 2-9.

$$\rho\left(\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z}\right) = -\frac{\partial p}{\partial x} + \frac{\partial \tau_{xx}}{\partial x} + \frac{\partial \tau_{xy}}{\partial y} + \frac{\partial \tau_{xz}}{\partial z} + S_M$$

\[ (2-9) \]

Where; $\tau_{ij} = \mu \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}\right)$ for a Newtonian fluid

The solution of the momentum transport equation relies on prior knowledge of the viscous characteristics of sludge, i.e., the relationship between shear stress, $\tau$, and velocity gradients, and can be obtained by rheological analysis of the fluid.

The equation of state for density, $\rho = \rho$(pressure, temperature), closes the system of 3D mass and momentum equations. In incompressible fluids, density is constant. Constant temperature is also assumed. Therefore the equation of state simplifies to $\rho = \rho$(specified).

In systems with multiple phases, each additional phase requires an additional set of conservation equations. A two fluid (Eulerian-Eulerian) multiphase model is the recommended approach when volume fractions of the dispersed phase exceed 10% [3], which is expected in wastewater applications. The ANSYS CFX Inhomogeneous Particle Model is used here, which is based on the Eulerian-Eulerian approach, and is reviewed below. The momentum and continuity equations for multiphase flow are detailed in Equations 2-10 and 2-11, respectively, and are similar in form to the single phase equations, but contain phasic volume fraction terms and additional source terms due to interface momentum transfer. Interphase mass transfer is not included in the following equations.

$$\frac{\partial}{\partial t}(r_\alpha \rho_\alpha v_\alpha) + \nabla \cdot (r_\alpha (\rho_\alpha U_\alpha \otimes v_\alpha)) = -r_\alpha \nabla p_\alpha + \nabla \cdot \left( r_\alpha \mu_\alpha (\nabla v_\alpha + (\nabla v_\alpha)^T) \right) + S_{M\alpha} + M_\alpha$$

\[ (2-10) \]

Where;

- $r_\alpha$ = volume fraction of phase $\alpha$
- $\rho_\alpha$ = density of phase $\alpha$
- $v_\alpha$ = phase $\alpha$ velocity vector
- $p_\alpha$ = pressure (phase $\alpha$)
- $\mu_\alpha$ = phase $\alpha$ viscosity
- $S_{M\alpha}$ = momentum sources
- $M_\alpha$ = force acting on phase $\alpha$ due to interaction with other phases
\[
\frac{\partial}{\partial t} (r_\alpha \rho_\alpha) + \nabla \cdot (r_\alpha \rho_\alpha U_\alpha) = S_{MS\alpha}
\] (2-11)

Where:
\[S_{MS\alpha} = \text{mass source}\]

The volume conservation equation provides an additional constraint that the sum of the volume fractions is unity (Equation 2-12) and can be included in the continuity equation (2-13).

\[\sum_{\alpha=1}^{N_p} r_\alpha = 1\] (2-12)

\[\sum_\alpha \left( \frac{1}{\rho_\alpha} \left( \frac{\partial}{\partial t} (r_\alpha \rho_\alpha) + \nabla \cdot (r_\alpha \rho_\alpha U_\alpha) \right) \right) = S_{MS\alpha}\] (2-13)

The system of equations are closed by the pressure constraint which specifies the same pressure for all phases i.e. \(p_\alpha = p\).

### 2.2.3 Turbulence transport equation and turbulence models

Laminar flows are fully described by the mass and momentum equations above. Flows above a certain Reynolds number (Re), which is the ratio of inertial forces to viscous forces \((\rho v L/\mu)\), are classed as turbulent. The point of transition depends on the flow problem. For example in pipe flow, flow is laminar at \(Re < 2100\), transitional at \(Re\) between 2100 and 4000 and turbulent at \(Re > 4000\). In tanks with mixing, flow is laminar at \(Re < 10\), transitional at \(Re\) between 10 and 10000 and turbulent at \(Re > 10000\) [18]. Velocity and other flow properties in turbulent flow deviate from basic momentum conservation expectation through fluctuations around a mean [8]. Turbulent flow can be approximated by the direct numerical simulation (DNS) of the Navier-Stokes equations, but the variations in length and time scales typical of turbulent flows makes this approach computationally expensive and inefficient [19]. Turbulence can be solved empirically, but the results of these models are not transferable to different configurations. The most widely used approach to general turbulence modelling is the use of averaging processes on the Navier-Stokes equations, where mean and fluctuations within the equations are considered [8]. This is called **Reynolds Averaging**. Velocity can be broken into a mean (time-averaged) component and a fluctuating component (Equation 2-14) and velocity components, \(u\), \(v\) and \(w\), in mass and momentum Equations 2-7 and 2-9 can be replaced by mean velocity components, \(\bar{u}\), \(\bar{v}\) and \(\bar{w}\). Stress components, \(\tau_{ij}\), now containing averaged velocity components are called Reynolds stresses.
\[ \mathbf{v} = \bar{\mathbf{v}} + \mathbf{v}' \]

Where, \( \bar{\mathbf{v}} = \) mean velocity (bar represents time-averaging)
\( \mathbf{v}' = \) fluctuating velocity

Therefore, the actual details of the turbulent flow need not be considered, only its effect on the mean flow behaviour. Turbulence models are required to close the system of mean flow equations. Typical Reynolds-Stress models that utilise Reynolds Averaging, include zero equation, one equation, two equation, algebraic stress (ASM) and stress/flux (Reynolds Stress Model, RSM) models. The most complex of these is the RSM which contains 6 equations describing turbulent transport and which therefore prohibits its use. The ASM is a simplification of the RSM, where Reynolds stress transport equations are approximated as a set of algebraic equations. The ASM however is a less widely used and validated model.

The zero, one and two equation models use the eddy viscosity (or diffusivity) approximation, which assumes a relationship between viscous stresses and mean velocity gradients [9, 19]. The two equation, or the k-\( \varepsilon \) turbulence model, is the most widely validated of the turbulence models [8, 9], is considered an industry standard [9] and is therefore implemented in this study. The turbulence kinetic energy, \( k \), represents the variance of the velocity fluctuations and the turbulence energy dissipation, \( \varepsilon \), defines the rate at which these velocity fluctuations dissipate. Equations 2-15 and 2-16 describe the link between turbulence kinetic and energy dissipation and effective fluid viscosity.

\[
\mu_t = C_\mu \rho \frac{k^2}{\varepsilon} \quad (2-15)
\]
\[
\mu_{\text{eff}} = \mu_t + \mu \quad (2-16)
\]

Where;
\( k \) = kinetic energy
\( \varepsilon \) = dissipation rate of kinetic energy per unit mass
\( \mu_t \) = turbulence viscosity
\( C_\mu \) = empirical constant; default value in Table 2-1
\( \mu_{\text{eff}} \) = effective viscosity

The pressure term is also modified to account for turbulence effects for incompressible fluids (Equation 2-17)
\[
p' = p + \frac{2}{3} \rho k \quad (2-17)
\]
Therefore, $\mu_{\text{eff}}$ and $p'$ replace $\mu$ and $p$, respectively in the momentum Equation (2-9).

The values of $k$ and $\varepsilon$ are calculated via additional transport Equations (2-18 and 2-19):

\[
\frac{\partial (\rho k)}{\partial t} + \nabla \cdot (\rho \mathbf{v} k) = \nabla \cdot \left[ \left( \mu + \frac{\mu_t}{\sigma_k} \right) \nabla k \right] + P_k - \rho \varepsilon \tag{2-18}
\]

\[
\frac{\partial (\rho \varepsilon)}{\partial t} + \nabla \cdot (\rho \mathbf{v} \varepsilon) = \nabla \cdot \left[ \left( \mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \nabla \varepsilon \right] + \frac{\varepsilon}{k} (C_{\varepsilon 1} P_k - C_{\varepsilon 2} \rho \varepsilon) \tag{2-19}
\]

Where:

$C_{\varepsilon 1}$, $C_{\varepsilon 2}$, $\sigma_k$ and $\sigma_\varepsilon$ are empirically obtained fitting constants (see Table 2-1 for default values)

$P_k = \text{turbulence production due to viscous forces} = \mu_t \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \frac{\partial u_i}{\partial x_j}$ for incompressible flows

Turbulence due to buoyancy, which is driven by density differences in the fluid, is not included in the above equations. In addition to the numerical difficulties encountered in solving buoyancy turbulence [20], turbulence due to density differences will be minimal compared with convection turbulence and hence is not considered in this study.

Default values of the empirical turbulence constants $C_\mu$, $C_{\varepsilon 1}$, $C_{\varepsilon 2}$, $\sigma_k$ and $\sigma_\varepsilon$ are detailed in Table 2-1, as described by the ANSYS CFX manual which are in accordance with the standard values provided by Rodi (1993) [19]. These constants were obtained by data fitting a wide range of turbulent flows against the $k$-$\varepsilon$ model [8].

**Table 2-1: Default values of turbulence constants**

<table>
<thead>
<tr>
<th>Constant</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_\mu$</td>
<td>0.09</td>
</tr>
<tr>
<td>$C_{\varepsilon 1}$</td>
<td>1.44</td>
</tr>
<tr>
<td>$C_{\varepsilon 2}$</td>
<td>1.92</td>
</tr>
<tr>
<td>$\sigma_k$</td>
<td>1.0</td>
</tr>
<tr>
<td>$\sigma_\varepsilon$</td>
<td>1.3</td>
</tr>
</tbody>
</table>
In multiphase problems the turbulence equations for the continuous liquid phase are modified to include liquid phase volume fraction terms and are similar in form to the single phase equations. Turbulence in the dispersed gas phase is linked to turbulence in the liquid phase via Equation 2-20.

\[
\nu_{t,G} = \frac{\nu_{t,L}}{Pr} \Rightarrow \mu_{t,G} = \frac{\rho_G \mu_{t,L}}{\rho_L Pr}
\]  

(2-20)

Where;

\( \nu_{t,G} \) = kinematic turbulent viscosity of the gas phase
\( \nu_{t,L} \) = kinematic turbulent viscosity of the liquid phase
\( \mu_{t,G} \) = dynamic turbulent viscosity of the gas phase
\( \mu_{t,L} \) = dynamic turbulent viscosity of the liquid phase
\( Pr \) = turbulent Prandtl number
\( \rho_G \) = density of gas
\( \rho_L \) = density of liquid

The turbulent Prandtl number is a ratio of the kinematic turbulent viscosities of the gas and liquid phases and is kept at the default value of 1.

In bubbly flows, an additional turbulence is generated on the continuous phase due to the motion of the bubbles. Therefore, turbulent velocity fluctuations can be divided into two components; a liquid turbulence independent of the bubble (shear induced) and a particle-induced turbulence due to bubble agitation [21], as described in Equation 2-21.

\[
\mu_{t,L} = \mu_{t,S} + \mu_{t,P}
\]

(2-21)

Where;

\( \mu_{t,L} \) = turbulent viscosity of the liquid phase
\( \mu_{t,S} \) = shear-induced turbulent viscosity
\( \mu_{t,P} \) = particle-induced turbulent viscosity

\( \mu_{t,P} \), also called Sato Enhanced Eddy Viscosity after the author, is described by Equation 2-22.

\[
\mu_{t,P} = 0.6 \rho_L r_G d_B |\mathbf{v}_G - \mathbf{v}_L|
\]

(2-22)

Where;

\( d_B \) = diameter of the bubble
Figure 2-1: Near wall flow velocity profile

Turbulent flows near solid walls are considerably different from free turbulent flows due to the no-slip condition imposed by solid boundaries. Near wall turbulence can be broken into two conceptual layers: a thin viscous sublayer immediately adjacent to the wall, followed by a turbulent region (Figure 2-1). Velocity in the viscous sublayer increases linearly with distance from the wall. After a certain distance from the wall, flow transitions into a turbulent region and velocity profile is logarithmic. To fully resolve near wall flow in turbulent flow situations, the number of mesh layers near the wall (inflation layers) needs to be very large, which introduces numerical issues around scaling. Wall functions are instead used to simplify near wall calculations in turbulent flow and make use of the consistent relationship between non-dimensional velocity, \( u^+ \) and non-dimensional distance \( y^+ \) (Figure 2-2). This relationship was determined from a series of experimental analyses conducted by Schlichting (1979) [22].
Assuming flow is in the x-direction, non-dimensional $y^+$ is related to the distance of the first mesh cell from the wall, $\Delta y_1$ (shown in Figure 2-2) by Equation 2-23.

$$y^+ = \frac{\Delta y_1}{v} \sqrt{\frac{\tau_w}{\rho}}$$  \hspace{1cm} (2-23)

Where:

$\tau_w$ = wall shear stress
$v$ = kinematic viscosity

Non-dimensional $u^+$ is the ratio of mean flow velocity, $\bar{u}$, to skin friction velocity, $u_\tau$ (Equation 2-24)

$$u^+ = \frac{\bar{u}}{u_\tau}$$  \hspace{1cm} (2-24)

Skin friction velocity is related to wall shear stress by Equation 2-25.

$$u_\tau = \sqrt{\frac{\tau_w}{\rho}}$$  \hspace{1cm} (2-25)

Below $y^+$ of 11, near wall flow is taken to be laminar (viscous-sublayer). Here, $u^+$ is equal to $y^+$ and wall shear stress is entirely viscous in origin (Equations 2-26 and 2-27).

$$u^+ = y^+$$  \hspace{1cm} (2-26)

$$\tau_w = \mu \frac{\bar{u}}{\Delta y_1}$$  \hspace{1cm} (2-27)
Above $y^+$ of 11, near wall flow is in the logarithmic region and wall functions for wall shear stress (via skin friction velocity) and turbulence parameters, $k$ and $\varepsilon$ are used (Equations 2-28 to 2-30).

$$u^+ = \frac{1}{\kappa} \ln (E y^+) \quad (2-28)$$

$$k = \frac{u_t^2}{\sqrt{C_m}} \quad (2-29)$$

$$\varepsilon = \frac{u_t^3}{k \delta y_1} \quad (2-30)$$

Where;

- $\kappa$ = von Karman’s constant $\approx 0.4$ for smooth walls
- $E$ = additive constant $\approx 9.8$ for smooth walls

ANSYS CFX utilises a ‘scalable’ wall function for the k-\varepsilon turbulence model which forces a minimum $y^+$ of 11. This in turn forces the use of wall functions for even arbitrarily fine meshes. The main assumption in a scalable wall function approach is that the profile of $u^+$ from the freestream to the wall follows the empirically obtained profile in Figure 2-2. To maintain consistent wall shear calculations in the scalable wall function approach, the first cell height should be monitored and adjusted for each flow scenario to maintain a constant $y^+$ so that it resides in the user’s desired region of Figure 2-2. In this study, $y^+$ was maintained in the transition region (around 11) in all turbulent flow scenarios.

### 2.2.4 Interphase (gas-liquid) momentum transfer (gas-liquid)

The system of phasic conservation equations are linked by interphase mass and momentum transfer equations. Interphase momentum transfer occurs due to the interface forces acting on each phase as a result of interaction with other phases, therefore total interface force $M_\alpha$ is the sum of interphase momentum transfer between phases, $M_{\alpha\beta}$ (when $\alpha \neq \beta$). Drag and lift are the most important contributing forces to the total interface force, and are hence considered the main drivers of interphase momentum transfer [23].

A variety of gas-liquid interphase models are available in commercial CFD codes. Yamoah, et al, 2015 [24], tested the numerical predictions of a number of these models against experimental results of vertical gas-liquid two phase flow in a pipe and found that the best agreement was obtained with the use of the Grace drag coefficient model, Tomiyama lift coefficient model, Antal’s wall lubrication force model and Favre averaged turbulent dispersion force model. These models
were hence chosen in two-phase gas-liquid modelling in this thesis, with the exception of the wall lubrication force model. The wall lubrication force model captures the tendency of bubbles to concentrate in the near-wall region in bubbly pipe flows and bubble columns where wall-effects on bulk flow are important. The effect of walls in bubbly flow is poorly understood [25] and wall lubrication force models can in some cases cause inaccuracies in near wall predictions of bubble flow [26]. In addition, very fine near-wall meshes are required to fully resolve wall lubrication effects, which will require considerable computational effort. Therefore, taking these reasons into consideration, the wall lubrication force model was neglected.

**Drag force** is exerted on a particle by a moving fluid and is hence proportional to phasic slip velocity, \( u_L - u_B \). Equation 2-31 defines drag force on a bubble exerted by a liquid continuum [25]. The dimensionless drag coefficient, \( C_D \), takes into account total drag force due to form (pressure forces) and skin friction (viscous shear forces).

\[
D_F = C_D \left( \frac{\rho_L}{\mu} (u_L - u_B)^2 A_B \right)
\]  
(2-31)

Where:
- \( C_D \) = Drag coefficient
- \( D_F \) = Drag force
- \( u_L \) = Velocity of fluid
- \( u_B \) = Velocity of bubble
- \( \rho_L \) = liquid phase density
- \( A_B \) = cross-sectional area of the bubble = \( \frac{d_B^2 \pi}{4} \)

The drag coefficient of gas bubbles is highly dependent on bubble shape regimes as described in Figure 2-3 [27]. Bubble shape characterisation requires knowledge of the non-dimensional bubble Reynolds number, \( Re_B \), (Equation 2-32), Eotvos number, \( Eo \) (Equation 2-33) and Morton number, \( M \) (Equation 2-34) [27].

\[
Re_B = \frac{\rho_L |u_L - u_B|d_B}{\mu_L}
\]  
(2-32)

Where; \( d_B \) = bubble diameter

\[
Eo = \frac{g \Delta \rho d_B^2}{\sigma}
\]  
(2-33)
Where; \( \sigma \) = surface tension coefficient
\( \Delta \rho \) = density difference between the two phases
\( d_B \) = diameter of the bubble
\( g \) = gravitational acceleration
\( \sigma \) = surface tension coefficient

\[
M = \frac{g \mu_1 \Delta \rho}{\rho^2 \sigma^3}
\]  

(2-34)

Figure 2-3: Bubble shape regimes in unhindered gravitational motion through liquids. From Clift et al. (1978) Bubbles, Drops and Particles [27].
Drag models for the determination of the drag coefficient are derived primarily from experimental data. At small bubble Reynolds numbers (viscous regime), the particles are roughly spherical and a Schiller-Naumann drag model [28] is utilised (Equation 2-35).

\[
C_D(\text{spherical}) = \frac{24}{Re_B} \left(1 + 0.15Re_B^{0.687}\right)
\]  

(2-35)

In intermediate regimes (distorted/ellipsoidal bubble regime), the drag coefficient becomes independent of Reynolds number and dependant on the Eotvos number. The Grace drag model is chosen which is described by the set of Equations 2-36 to 2-39 [27].

\[
C_D(\text{ellipsoidal}) = \frac{4gd_B \Delta \rho}{3 U_T^2 \rho_L}
\]  

(2-36)

\[
U_T = \text{Terminal velocity} = \frac{\mu_L}{\rho_L d_B} M^{-0.149} (J - 0.857)
\]  

(2-37)

\[
J = \begin{cases} 
0.94H^{0.757}, & 2 < H \leq 59.3 \\
3.42H^{0.441}, & H > 59.3
\end{cases}
\]  

(2-38)

\[
H = \frac{4}{3} \text{EoM}^{-0.149} \left(\frac{\mu_L}{\mu_{\text{ref}}}\right)^{-0.14}
\]  

(2-39)

Where;

\(\mu_{\text{ref}}\) = reference viscosity, generally of water at 25°C

At large bubble Reynolds numbers, (inertial/spherical cap regime), the drag coefficient approaches a constant value of 8/3 [25] (Equation 2-40).

\[
C_D(\text{spherical cap}) = \frac{8}{3}
\]  

(2-40)

A bubble experiences a shear-induced lift force perpendicular to the direction of the relative motion of the liquid and particle phases. Equation 2-41 describes the lift force acting on a bubble in the presence of a rotational liquid phase (also called the shear-induced lift force model) [25, 29, 30].

\[
L_F = r_B \rho_L C_l (U_B - U_L) \times \omega_L
\]  

(2-41)

Where;

\(L_F\) = Lift force

\(C_l\) = Lift coefficient

\(\omega_L\) = curl \(U_L\)
The Tomiyama lift model [30] is applicable to deformable bubbles in the ellipsoidal and spherical cap regimes and is expressed in Equation 2-42.

\[
C_L = \begin{cases} 
\min\left[0.288\tanh(0.121Re_B, f(Eo'))\right], & Eo' \leq 4 \\
0.00105Eo'^3 - 0.0159Eo'^2 - 0.0204Eo' + 0.474, & 4 < Eo' \leq 10 \\
-0.27, & 10 < Eo' 
\end{cases}
\] (2-42)

Where;
\[Eo' = \text{modified } Eo = \frac{g(\rho_L - \rho_B)d_H^2}{\sigma}\]
\[d_H = \text{long axis diameter of deformable bubble} = d_B(1 + 0.163Eo^{0.757})^{1/3}\]

**Turbulent dispersion force** is an additional interphase force which results in the dispersion of particulate phases from high to low volume fractions due to turbulent fluctuations. The Favre-averaged drag model, developed by Burns et al, 2004 [26], is utilised here. This model is primarily derived using the concepts of Favre-averaging and eddy-diffusivity hypothesis.

Favre-averaging or mass-weighted averaging of a scalar quantity, \(\varphi\), is defined by Equation 2-43 (assuming no density fluctuations).

\[
\overline{\varphi}_\alpha = \frac{\bar{r}_\alpha \varphi_\alpha}{\bar{r}_\alpha} \to \bar{r}_\alpha \overline{\varphi}_\alpha = \bar{r}_\alpha \varphi_\alpha
\] (2-43)

Where;
\(\overline{\varphi}_\alpha\) = Favre-averaged scalar quantity
\(\bar{r}_\alpha\) = mean (time averaged) volume-fraction of phase \(\alpha\) (overbar denotes time-averaging)
\(\overline{\varphi}_\alpha\) = mean (time averaged) component of the scalar quantity

If a quantity, \(\varphi\), comprises a mean (time-averaged) component and a fluctuating component \((\varphi = \overline{\varphi} + \varphi')\) then the relationship between time-averaged and Favre-averaged equations can be derived (Equation 2-44.) This equation now includes the fluctuating components of \(\varphi\).

\[
\bar{r}_\alpha \overline{\varphi}_\alpha = \bar{r}_\alpha \overline{\varphi}_\alpha = (\bar{r}_\alpha + \bar{r}'_\alpha)(\overline{\varphi}_\alpha + \varphi'_\alpha) \to \overline{\varphi}_\alpha = \overline{\varphi}_\alpha + \frac{\bar{r}_\alpha \varphi'_\alpha}{\bar{r}_\alpha}
\] (2-44)

Substituting velocity, \(v\), in \(\varphi\) the relationship between Favre-averaged and time-averaged velocities is obtained (Equation 2-45).
\[ \nabla \underline{\alpha} = \underline{v}_\alpha + \frac{r_{\alpha} v_{\alpha}}{r_{\alpha}} \]  

(2-45)

The term \( r_{\alpha} v_{\alpha} \) describes the dispersion of phasic volume fractions due to velocity fluctuations and is hence fundamental to turbulent dispersion.

In the eddy-diffusivity hypothesis, turbulent transport of a quantity is directly proportional to the gradient of the mean value of the transported quantity and is described by Equation 2-46:

\[ \nabla' \phi' = -\frac{v_t}{Pr} \nabla \phi \]  

(2-46)

Where:

- \( v_t \): turbulent kinematic viscosity
- \( Pr \): turbulent Prandtl number (default value = 1)
- \( \phi \): mean component of scalar quantity
- \( \phi' \): fluctuating component of scalar quantity

Hence, substitute \( \phi \) with volume fraction, \( r_{\alpha} \) to give the eddy diffusivity equation for turbulent transport of volume fraction of phase \( \alpha \) (Equation 2-47).

\[ \nabla' r_{\alpha}' = -\frac{v_t}{Pr} \nabla \overline{r_{\alpha}} \]  

(2-47)

Introducing the general form of interphase drag force in Equation 2-48 which describes drag force experienced by phase \( \alpha \) due to the motion of phase \( \beta \).

\[ D_{F\alpha} = C_{\alpha \beta} (v_\beta - v_\alpha) \]  

(2-48)

Where:

- \( D_{F\alpha} \): Drag force on phase \( \alpha \)
- \( C_{\alpha \beta} \): momentum transfer coefficient
- \( (v_\beta - v_\alpha) \): interphase slip velocity

Rearrange Equation 2-47 to make \( \nabla \overline{r_{\alpha}} \) the subject and substitute into the velocity terms in Equation 2-48 to derive the general form of the Favre-averaged drag model (Equation 2-49).
F_{TD,\alpha} = -F_{TD,\beta} = C_{\alpha\beta} \frac{v_{\alpha}}{Pr_\alpha} \left( \frac{v_{r_{\beta}}}{r_{\beta}} - \frac{v_{r_{\alpha}}}{r_{\alpha}} \right) \quad (2-49)

Where;

\( F_{TD} \) = Turbulent dispersion force

\( C_{\alpha\beta} \) is the momentum transfer coefficient for interphase drag force and is related to the drag coefficient, \( C_D \). For example, in a dispersed two phase-flow with continuous phase, \( \alpha \) and dispersed phase, \( \beta \) with a (Sauter) mean diameter of \( d_\beta \), the relationship between \( C_{\alpha\beta} \) and \( C_D \) is described in Equation 2-50.

\[
C_{\alpha\beta} = \frac{3}{4} C_D \frac{r_{\beta} \rho_\alpha}{d_\beta} |v_\beta - v_\alpha| \quad (2-50)
\]

Hence, the Favre averaged drag model is linked to the choice of the interphase drag model. This model was validated against bubbly flows [24, 26] with good agreement between predicted and measured results. Burns et al, 2004 [26] found better agreement with the Favre averaged drag model when compared with other turbulent dispersion models (such as the Lopez de Bertadano and Carrica models).

### 2.2.5 Bubble size

Approximate estimations of bubble size are utilised in this study. These empirically obtained approximations relate bubble size with surface tension of the liquid, sparger orifice diameter, fluid viscosity and gas flow rate [31]. At low gas flow rate, bubble flow can be modelled by the formation of a bubble at a single circular orifice and is dependent on orifice diameter (Equation 2-51).

\[
d_B = \left( \frac{6D\gamma}{g \Delta \rho} \right)^{1/3} \quad (2-51)
\]

Where;

\( d_B \) = bubble diameter [m]
\( D \) = orifice diameter [m]
\( g = 9.81 \) [m/s\(^2\)]
\( \Delta \rho \) = density difference between liquid and gas phase =\( |\rho_L - \rho_B| \) [kg/m\(^3\)]
At high gas flow rate the bubble diameter is less influenced by orifice diameter and more dependent on liquid viscosity (laminar) and gas flow rate (turbulent), in accordance with Equations 2-52 and 2-53.

In laminar conditions:

\[ d_B = \left( \frac{108 V_G \mu}{\pi g \Delta \rho} \right) \]  

(2-52)

In turbulent conditions:

\[ d_B = \left( \frac{72 V_G^2 \rho}{\pi^2 g \Delta \rho} \right)^{1/3} \]  

(2-53)

Where:

\[ V_G = \text{gas flow rate [m}^3/\text{s]} \]
\[ \mu = \text{liquid viscosity [Pa s]} \]

It should be noted that bubble break up and coalescence is not considered in this study.

### 2.2.6 Density calculations

Effective density of liquid is calculated using an ideal mixture model (Equation 2-54).

\[ \rho = \left[ \rho_p \times \frac{TS}{\rho_w} \right] + \left[ \rho_w \times \left( 1 - \frac{TS}{\rho_w} \right) \right] \]  

(2-54)

Where:

\[ \rho_p = \text{density of particle [kg/m}^3] \]
\[ TS = \text{total solids concentration [kg/m}^3] \]
\[ \rho_w = \text{density of water = 998 kg/m}^3 \text{ at 20°C} \]

The density of suspended floc was assumed to be 1100 kg/m\(^3\) in accordance with measurements in anaerobically digested activated sludge by Sears et al, 2006 [32].
2.2.7 Buoyancy

Buoyancy is driven by density differences in the fluid which arise due to local temperature variations or in fluids with multiple components and/or phases. Therefore, in a single phase flow with constant temperature and density, there is no buoyance force. In multiphase flow, buoyancy is taken as an additional source term in the momentum equations, and is simply calculated using a density difference between the phases, $\rho_L - \rho_B$.

2.2.8 Modelling of solids phase

The solids phase is modelled using the Algebraic Slip Model which is a simplified multiphase model that treats the solid phase as a pseudo-solute with gravity and buoyancy separation simulated by a slip function. In this study, a slip velocity is specified based on empirically obtained settling velocity measurements. This approach is particularly useful in WW modelling as sludge particles behave differently to classical solid particles. Sludge solids follow a hindered settling behaviour, where settling velocity decreases as concentration of solids increases. Empirical settling models are commonly applied in WW studies and can be used to describe slip velocity in the CFD model. Commonly employed empirical settling models are summarised in Table 2-2.

<table>
<thead>
<tr>
<th>Model</th>
<th>Equation</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$v = v_0 - k[TS]$</td>
<td>[33, 34]</td>
</tr>
<tr>
<td>2</td>
<td>$v = v_0 (1 - k[TS])^{4.65}$</td>
<td>[35]</td>
</tr>
<tr>
<td>3</td>
<td>$v = v_0 e^{-k[TS]}$</td>
<td>[36]</td>
</tr>
<tr>
<td>4</td>
<td>$v = v_0 e^{-k_h[TS]} - v_0 e^{-k_p[TS]}$</td>
<td>[37]</td>
</tr>
<tr>
<td>5</td>
<td>$v = v_0 [TS]^{-n}$</td>
<td>[38, 39]</td>
</tr>
</tbody>
</table>

$v$ = Settling velocity (m/s); $v_0$ = terminal settling velocity (m/s); $k$, $k_h$, $k_p$ = settling constants (m$^3$/kg) and $TS$ or $TS^*$ = solids concentration [kg/m$^3$]
2.2.9 Rheological models

The mechanical properties of wastewater (i.e. its fluid dynamic characteristics) can be described by its rheological characteristics [40, 41]. Rheology is defined as the viscous characteristics of a sludge and relates the shear stress experienced by the fluid at a range of shear rates [42]. The modelling of viscous flow in the reactor and through the membrane requires a viscosity model, which describes changes in viscosity with applied shear and at different temperatures and solids concentrations. Additionally, fouling of the membrane, which is highly dependent on process hydrodynamics, requires knowledge of sludge viscosity for the determination of the hydraulic regime and transport phenomena near the membrane [43]. Meng et al [44], established a link between membrane fouling and sludge viscosity, the latter being directly dependent on sludge solids concentration. Dilute wastewaters generally behave in a Newtonian fashion [45], i.e. the relationship between shear stress, $\tau$, and shear strain, $\gamma$, is linear (fluid viscosity, $\mu$, is constant).

Concentrated sludge however is typically non-Newtonian and can display one of three behaviours (Figure 2-4):

(i) Pseudoplastic, where fluid viscosity decreases as shear strain increases;

(ii) Bingham, which exhibits a yield stress that has to be overcome before the fluid can flow; and

(iii) Yield pseudoplastic which exhibits both non-linear behaviour and a yield stress.

![Figure 2-4: Newtonian and non-Newtonian fluid behaviours](image)
2.2.10 Solving the fluid mechanics equations

The governing equations for fluid flow are complex, non-linear partial differential equations (PDEs) which, at the scale and geometry applied need to be numerically solved. Numerical solution of PDEs requires discretisation of the PDEs over a spatially discretised domain (grid). ANSYS CFX, a commercially developed CFD platform is utilised in this work. ANSYS CFX utilises the finite volume method of evaluating PDEs. In this method, the spatial domain (i.e. the fluid volume within the tank or reactor) is discretised or meshed into finite control volumes over which fluid flow is conserved. Unstructured grids are required in complex and curved geometries, where mesh elements can easily be concentrated as required. Three-dimensional unstructured grids most commonly comprise a mixture of tetrahedral, hexahedral, pyramid or triangular prism (wedge) shaped elements. Triangular prisms are most commonly employed for boundary layers in near wall meshing. During the mesh generation stage, mesh quality can be monitored to minimise sharp internal angles via monitoring orthogonality and skewness. Mesh quality is considered ‘good’ below skewness of 0.8 and above orthogonality of 0.2.

ANSYS CFX discretises the PDEs of fluid flow using a vertex-centred method. Here, nodes are placed on the vertices of the grid and control volumes are formed by connecting centroids of the element and midpoints of the edges. Figure 2-5 illustrates the structure of typical mesh elements as utilised by ANSYS CFX (shown in two-dimensions for simplicity).

![Figure 2-5: Structure of mesh elements](image-url)
In ANSYS CFX, all solution variables and fluid properties are stored at mesh nodes. Surface integrals of the governing equations, such as the convective and diffusive flux terms, are discretised at the integration points of the element. Volume integrals such as the transient term and volume sources are discretised within each element sector. Surface integrals (fluxes) at an integration point are equal and opposite for control volumes adjacent to the integration point; and are hence locally conserved.

Discrete approximations of the PDEs are based on boundary fluxes with higher orders approximated by Taylor series approximations and order accuracy determined by the truncation of the series. Central differencing estimates the value of a point based on interpolations of its neighbouring nodes and hence values at a node are equally affected by its neighbouring nodes. Treatment of convective fluxes can be done via upwind differencing (since this is first order) instead since the upwind node has the largest impact on flux into the control volume. A hybrid differencing scheme switches between central and upwind differencing based on the ratio of convection to diffusion; i.e the Peclet number. Higher order schemes that use polynomial functions such as the quadratic upstream interpolation for convective kinetics (QUICK) increase order accuracy for convection problems but face stability issues in high Peclet number flows. High Resolution schemes minimise oscillations in the solution while maintaining second or higher order accuracy by introducing a blending function that ensures that the calculated value of the convective flux at an integration point does not overshoot the maximum value and undershoot the minimum value of the flux at neighbouring points.

In structured grids, the differencing schemes mentioned above can use straightforward linear interpolation. However, in unstructured grids, interpolation is dependent on the shape of the element. Therefore, the calculation of gradients at integration points require the use of interpolation shape functions for hexahedral, tetrahedral, wedge and pyramid shaped elements.

In this study, a second order high resolution scheme was used for the convection term. Turbulence terms are discretised using a first order upwind scheme to minimise time to convergence. Pressure and diffusion terms are evaluated based on their gradients at integration points and the difference equations are hence dependent on the local element shape function.

ANSYS CFX uses a coupled solver which combines pressure and velocity in a single solution matrix. Coupling is hence retained at the equation level. This is in contrast to segregated algorithms where pressure and velocity are separated and coupled via a pressure correction equation. Coupled solvers are generally more robust and efficient than segregated solvers and require less time for convergence; but have large memory requirements. The algorithm uses a fully implicit solver,
where the solution is advanced in time (or false time in steady state where the time step is used as an acceleration parameter). Each time step involves two numerically intensive operations; coefficient generation and equation solution. In coefficient generation, the discretised, non-linear mass and momentum equations are linearised into a matrix form $[A][\emptyset] = [b]$; where $[A]$ is the coefficient matrix, $[\emptyset]$ is the solution matrix and $[b]$ comprises the constant vector. In the second equation solution step, the linearised equations are solved using an Algebraic Multigrid method. The multigrid technique was developed to enable more rapid convergence to an approximate solution at a coarse scale (or considering limited nodes).

Hence error in flow calculations can be controlled by using higher order discretisation schemes such as the high resolution scheme, reducing mesh size in regions of rapid solution variation and managing shape functions of the mesh element such that mesh quality is sufficiently high.

General CFD workflow includes a four step process; i) setting up the physical fluid geometry, ii) meshing the fluid; iii) describing the flow problem including initial and boundary conditions and iv) solution and post-processing.

2.3 OVERVIEW OF CFD SIMULATIONS

2.3.1 CFD shear validation experiments

The main objective of CFD modelling in this thesis is prediction of membrane shear to enable further model based prediction of membrane fouling. To test the validity of the CFD model for shear estimation, the model was applied and validated against an experimental study (Section 2.5.2) obtained from literature [14] that measured shear on a submerged Teflon fibre.

In this study, the fibre was submerged in a bench-scale tank with a 16 L working volume. The tank had a diameter of 14.2 cm and a height of 110 cm (height of water level at 101 cm). The fibre was placed within a cylindrical baffle (6.3 cm in diameter and 80 cm in height). The fibre had a diameter of 2 mm and a height of 42 cm. Shear on the fibre was measured using electrochemical shear probes installed at three points (marked A, B and C in Figures 2-6a and 2-6b).
Shear was measured with two different sparging configurations (outer-ring sparger shown in Figure 2-6a and inner sparger shown in Figure 2-6b) and at three different sparge rates that maintained bulk crossflow velocities at 0.2, 0.3 and 0.4 m/s in both sparger configurations. Nitrogen was used as the sparger gas. Experimental conditions simulated in the CFD model are summarised in Table 2-3.

Table 2-3: Summary of experimental conditions

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Crossflow velocity (m/s)</th>
<th>Sparge rate [L/min]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.2</td>
<td>3.3</td>
</tr>
<tr>
<td>1. Outer ring sparger</td>
<td>0.3</td>
<td>6.1</td>
</tr>
<tr>
<td></td>
<td>0.4</td>
<td>11.4</td>
</tr>
<tr>
<td></td>
<td>0.2</td>
<td>2.9</td>
</tr>
<tr>
<td>2. Inner sparger</td>
<td>0.3</td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td>0.4</td>
<td>7.3</td>
</tr>
</tbody>
</table>

Both sparger configurations were meshed into approximately 960,000 elements. A two-phase (liquid-gas) model was employed for both configurations. A k-ε turbulence model and interphase momentum models as described in Sections 2.2.3 and 2.2.4 were used. A mean bubble size of 3 mm was assumed based on bubble size estimations using Equations 2-52 and 2-53. The viscosity of
water at 25°C was assumed to equal 0.0009 Pa.s. The density of nitrogen gas was taken as 1.165 kg/m³ at 25°C. Convergence is defined as residuals of $1 \times 10^{-3}$ and when monitored parameters (volume-averaged fluid velocity and area-averaged membrane shear) reached constant values.

### 2.3.2 Mesh independence

Two phase CFD simulations (water and 3.5 L/min gas flow rate) were conducted on a pilot scale AnMBR geometry (Figure 2-7) meshed into 342,000 elements and 1,324,000 elements and the velocity and shear results compared. The solution was considered mesh independent if no difference in these monitored values were observed with mesh resolution.

Figure 2-7: Pilot AnMBR layout
2.3.3 Two vs. three phase CFD

The purpose of three phase CFD analysis was to determine if solids settling has an effect on reactor hydrodynamics in the pilot AnMBR. Modelling of solids behaviour provides additional mechanisms including linking of viscosity and density to concentration but is computationally expensive and usually ignored as solids settling is overcome by positive buoyancy forces. In cases where mixing is not sufficient, solids separation from the bulk liquid will be observed, and hence the fluid can no longer be approximated by a homogenous liquid phase. Solids settling rate is typically fastest at low solids concentration, and hindering reduces velocity at higher concentrations. Therefore, a three phase study was conducted at low solids concentration (2 g/L) and at a low sparge rate (3.5 L/min), i.e. conditions where solids separation will be expected, and the results compared with a two phase approximation to determine if solids settling greatly impacts model outcomes and if three phase modelling is required in future studies.

CFD analyses were conducted on a geometry based on an existing pilot-scale AnMBR. The AnMBR is a 200 L bioreactor (0.47 m diameter by 0.778 m wetted height) (Figure 2-7) with a vertical mounted submerged hollow fibre membrane (ZW10 from ZENON Environmental Inc.).

For dilute Newtonian liquids, Equation 2-55 [46] was used to estimate viscosity.

\[
\mu = \frac{0.33X+2.3}{1+0.0337X+0.000221T^2} \tag{2-55}
\]

Where;
- \(\mu\) = sludge viscosity [Pa.s]
- TS = Total solids concentration, [g/L]
- T = Temperature, [°C]

For higher TS concentrations, a Casson (yield pseudoplastic) model based on Pevere et al’s (2007) [47] measured correlations on anaerobic sludge up to 22 gTS/L (Equations 2-56 to 2-58) were adopted.

\[
\sqrt{\mu} = \sqrt{\tau_Y} + \sqrt{K} \tag{2-56}
\]

\[
\tau_Y = 0.023e^{0.06\text{TS}} \tag{2-57}
\]

\[
K = 0.0005e^{0.08\text{TS}} \tag{2-58}
\]
Where;

\[ \tau_Y = \text{yield stress} \ [\text{Pa}] \]
\[ \gamma = \text{Shear strain rate} \ [\text{s}^{-1}] \]
\[ K = \text{viscosity consistency} \ [\text{Pa.S}] \]

At dilute concentrations of 2 gTS/L, the behaviour of sludge will follow a Newtonian behaviour [46]. In a two-phase approximation, constant viscosity was assumed throughout the reactor and a viscosity of 0.0015 Pas at 25°C was used based on Equation 2-55. In three-phase with solids settling, the TS concentrations will vary inside the reactor, and hence rheology was linked with local TS concentrations (Figure 2-8). Tank zones with TS below 5 gTS/L used the Newtonian correlation equation (2-55) while zones with greater than 5 gTS/L used the non-Newtonian Casson viscosity model and correlations (equations 2-56 to 2-58). Room temperature (25°C) was assumed in these scenarios.

![Viscosity relationships used in this study. Below 5 g/L, Newtonian rheology was assumed and Psoch and Schiewer (2008) viscosity-MLSS correlations were used. Above 5 g/L, non-Newtonian rheology was assumed, i.e. viscosity becomes shear rate dependent, and Casson model correlations (Pevere, 2006) were used.](image)

At a gas sparge rate of 3.5 L/min, the low gas rate equation (2-51) was used, giving a bubble size of 4 mm.
2.4 EXPERIMENTAL DETERMINATION OF SOLIDS SETTLING VELOCITY

This was used to obtain the settling velocity-concentration relationship. Zone settling tests are generally conducted to estimate settling velocity and to identify an appropriate settling model.

Zone settling tests were conducted as per the protocol described by White (1975) [48], and in accordance with APHA Standard Method #2710E [49]. A well-mixed sludge sample of a known TS concentration was placed in a 1 L measuring cylinder. An interface is formed separating sludge solids from relatively clear supernatant. The height of this interface was recorded at various times to produce a curve illustrating interface displacement with time. This curve is typically linear at first and later plateaus as the system changes to compression settling (as solids concentration at the interface rises) (Figure 2-9). The gradient of the linear part is the settling velocity, v for the given solids concentration. This test is repeated with samples of different values of TS concentrations, to determine the relationship between v and TS.

![Figure 2-9: Zone settling test](image)

2.5 RESULTS AND DISCUSSION

2.5.1 Solids settling velocity

The relationship between settling velocity, v and TS concentration obtained from batch settling tests in AnMBR sludge (treating red stream from a slaughterhouse wastewater plant) is shown in Figure 2-10 (duplicate measurements shown). This relationship is linear (adjusted R-squared value of 0.99) and hence follows Model 1 in Table 2-2. The intercept, \(v_0\) and slope, k is estimated at \((1.2 \pm 0.21) \times 10^{-3}\) m/s and \((8.63 \pm 2.03) \times 10^{-5}\) m\(^3\)/kg, respectively.
Figure 2-10: Settling velocity vs total solids concentration of sludge from the pilot scale AnMBR and sludge treating food processing wastewater.

Settling behaviour of an anaerobic sludge treating food processing wastewater [50] is shown in Figure 2-10 alongside the measured settling velocity of AnMBR sludge, for comparison. An exponential Vesilind model best fits the settling behaviour of food processing fed anaerobic sludge; with parameters, $v_0$ and $k$, estimated at $1.1 \times 10^{-3} \text{ m/s}$ and $0.368 \text{ m}^3/\text{kg}$, respectively. AnMBR sludge therefore has a similar terminal velocity to food processing WW sludge but possesses a higher degree of settleability. Sludge with higher settleability will require greater energy input to keep the sludge in suspension. The linear settling model for the AnMBR was hence implemented into the three phase CFD model, as it will provide a conservative model when deciding whether the full three phase model will be required for the rest of the study. This settling model was linked to local solids concentration via an algebraic equation of the form in Settling Model 1 (Table 2-2) substituting the experimentally obtained AnMBR settling parameters into $v_0$ and $k$. Above 14 gTS/L, settling velocity was fixed at $1 \times 10^{-6} \text{ m/s}$.

### 2.5.2 Model validation

Figures from the original study by Bèrubè et al (2006) [14] are adapted here for presentation purposes. Figure 2-11 shows typical time-variations in measured surface shear stress (shown here at crossflow velocity of 0.3 m/s). Measured shear in configuration 1 is relatively constant over time.
However, in configuration 2, measured shear values were highly variable; and in general higher than in configuration 1. In configuration 1, bubbles are kept outside the cylindrical baffle and hence flow along the Teflon fibre is effectively single-phase. In configuration 2, flow along the Teflon fibre is two-phase, and bubbly. In bubbly flow, two components to shear stress are identified; (i) localised high liquid velocities in a falling film that forms between a gas bubble and the wall surface and (ii) formation of turbulent eddies that are induced by a rising gas bubble. These phenomena have the effect of imparting a higher shear on the fibre in dual-phase crossflow (configuration 2) than in single-phase crossflow (configuration 1), and of increasing time-dependent instabilities in the flow which are captured by variations in dual-phase measurements (configuration 2). Shear stress values obtained from CFD simulations of both experimental configurations are superimposed on Figure 2-11 as red (configuration 1) and blue (configuration 2) lines, and are in good agreement with measured values.

Figure 2-11: Typical shear variations at 0.3 m/s crossflow velocity in single-phase (outer ring sparger configuration) and dual-phase (inner sparger configuration), adapted from Bérubè et al. (2006). Blue and red lines depict simulated values from CFD analyses conducted (this thesis).
Figure 2-12 summarises the time-averaged values and 90% confidence intervals of measured shear at the three probe locations in single-phase (Figure 2-12a) and dual-phase (Figure 2-12b) configurations. In Figures 2-12a and 2-12b, superimposed red lines mark the CFD simulated values of shear (area-averaged over the entire fibre) and are within the 90% confidence intervals of measured shear values.

This model validation exercise suggests that CFD modelling provides an objectively accurate estimate of wall shear and bulk shear forces, but that cyclic variation is not predicted due to complex phenomena not accounted for, possibly including mechanical coupling. This is not likely to materially influence suitability of the model to predict cake accumulation.
Figure 2-12: Surface shear distribution along the fibre surface in; (a) configuration 1 (outer ring sparger) and (b) configuration 2 (inner sparger). Error bars represent the 90% confidence interval of the measurements. Adapted from Bèrubè et al. (2006). Red lines depict simulated values from CFD analyses (conducted in this thesis) of both configurations.
2.5.3 Mesh independence

No variation in reactor averaged velocity or membrane averaged shear rate (Figure 2-13) was observed when the AnMBR mesh was refined from 342,000 elements to 1,324,000 elements; hence the coarser mesh was used in CFD simulations and the solution considered mesh independent.

![Figure 2-13: Mesh independence study](image)

2.5.4 Two vs. three phase modelling

Figure 2-14a shows that in a reactor with total initial concentration of 2 gTS/L and gas flow rate of 3.5 L/min, solids settling will take place and hence a spatial variation in TS will exist inside the reactor. Above the sparger, TS concentration is generally homogenous at approximately 2 g/L. The settling of solids below the sparger confirms the presence of stagnant zones. The dynamic viscosity in three phases varies with local TS concentration (Figure 2-14b). There is however little difference in profiles of bulk flow velocity (Figure 2-15a) and membrane shear (Figure 2-15b) between the two models (summarised in Table 2-4). This is attributed to the similar TS concentrations (2 g/L) and hence viscosity above the sparger in both scenarios.

It should be noted that the measured settling velocity of AnMBR sludge is higher than the literature reported settling velocity of anaerobic sludge treating food processing waste (as described earlier in section 2.5.1). Therefore, the negligible effect of solids settling on key flow variables (in conditions such as low TS and sparge rate where solids settling is expected to be the highest) suggests that the assumption of a homogenous liquid in a two-phase model is sufficient for future modelling studies.
of the AnMBR pilot. Simulation time for the three phase model (wall-clock time of 82 hours) is much higher than the two phase model (wall-clock time of 2 hours) (Table 2-4), therefore, a two-phase assumption will also have greater computationally efficiency.

Figure 2-14: Three-phase CFD simulation of 2 gTS/L at a sparge rate of 3.5 L/min. (a) variation of solids concentration within the tank and (b) local variation in viscosity
Figure 2-15: Two vs. three phase results; a) velocity and b) membrane shear
Table 2-4: Two vs. three phase key variable values and simulation time

<table>
<thead>
<tr>
<th></th>
<th>Volume averaged velocity [m/s]</th>
<th>Membrane area averaged shear rate [s⁻¹]</th>
<th>Wall clock hours [h]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Two phase</td>
<td>0.0277</td>
<td>6.53</td>
<td>1.57</td>
</tr>
<tr>
<td>Three phase (algebraic slip)</td>
<td>0.0281</td>
<td>6.72</td>
<td>81.8</td>
</tr>
</tbody>
</table>

2.6 FURTHER MODEL APPLICATIONS AND MODEL ASSUMPTIONS/LIMITATIONS

The validity of a CFD model for the prediction of membrane shear relies on suitable boundary layer meshing and appropriate empirical submodels. The chosen model approach provides membrane shear results which are in good agreement with measured shear, therefore providing confidence in the overall meshing and model approach for further pilot-scale AnMBR shear estimations. The CFD model links membrane shear with operating conditions, such as sparge rate and viscosity (total solids), as well as to internal reactor geometry, therefore enabling the study of these effects on shear during reactor design and optimisation. CFD shear can further be used for the prediction of cake accumulation on a membrane surface. The capability of the CFD model to provide multidimensional shear information will enable the prediction of non-uniform planar shear on membrane fouling. Chapter 4 discusses the development of a model for prediction of membrane fouling on planar membrane surfaces, such as flat sheet membranes, using CFD shear. Chapter 5 discusses treatment of CFD shear for multiple fibre membranes, such as hollow fibre membranes.

A major assumption here is that CFD model validation against lab-scale measurements is sufficient. Ideally, the model would be validated against larger full-scale MBR shear measurements, which will provide greater confidence in the real-world applicability of the CFD model. However; large-scale shear measurements are difficult as experimental flow visualisation techniques such as, the use of tracer dye clouds and particle image velocimetry, are difficult in opaque and transient wastewater flows. Acoustic Doppler Velocimetry (ADV) probes are often used for full-scale flow estimation but cannot be implemented in MBRs due to the tight spaces between membranes and membrane modules. Therefore simplified small-scale flow experiments are generally the only available option for shear measurement and hence CFD shear validation. In this thesis a pilot-scale (200 L) reactor was the focus of CFD model analyses, hence a full-scale validation exercise was considered unnecessary.
A major difference expected between small and larger scale MBRs is the effect of variable bubble size, which is not accounted for in the CFD model. A constant bubble size was assumed, with no bubble break-up and coalescence effects. Bubble size generally increases with height, due to bubble coalescence and reduction in static pressure. These effects are enhanced in taller, full-scale tanks. The effect of bubble size on shear is complex. In bubbly flow regimes (spherical to spherical-cap), smaller bubbles impart higher shear than larger bubbles at the same gas flowrate [51]. In turbulent flow in pipes and airlift reactors at high gas flowrates, bubble size can increase to a point where the flow regime is changed from bubbly flow to bubble-slug or turbulent churn flow, which impart high turbulent wall shear [52, 53]. Therefore neglecting the increase in bubble size may overestimate shear in a bubbly flow regime and underestimate shear in a bubble-slug or turbulent churn regime. These effects may be important in tall reactors containing long membranes, and will affect the upper parts of the membrane where bubble flow is fully developed.

In this thesis, it is assumed that for the the pilot-scale reactor (200 L), the effect of bubble size will not be significant as the reactor is relatively short compared to full scale installations. The pilot scale was generally operated at low gas flowrate (3.5 L/min) in dilute wastewater. High gas flowrates (30 L/min) were only utilised for concentrated sludge (20-35 gTS/L). Therefore, in both situations, conditions are not conducive for the development of significantly large bubbles or bubble slug flows, and hence a constant mean size assumption is considered sufficient. It is further assumed that non-Newtonian rheological correlations from lab-scale rheometer measurements sufficiently describe shear-viscosity relationships in a larger scale reactor with concentrated sludge.

Another phenomenon not accounted for in the CFD model is the effect on flow profiles of biogas produced by anaerobic biomass. It is assumed here that the impact of in-reactor produced biogas flow rates on membrane shear will be insignificant compared with that of the sparger gas flow rate.

2.7 CONCLUSIONS

The chosen CFD approach showed good agreement with experimental measurements of shear. Therefore, this approach provides objectively accurate estimations of wall shear, and will be applied in later CFD analyses for AnMBR design and optimisation. Minimal differences in two and three phase model outcomes suggest that a two-phase assumption will be sufficient in further analyses. It is assumed that model validation against lab-scale experiments is sufficient and that the validated approach will be applicable for larger scales. The major difference expected between small and larger scale installations is the effect of bubble coalescence and transient bubble regimes on
membrane shear. These effects are expected to be minimal in the pilot scale AnMBR compared with larger scale tanks. The effect on membrane shear of biologically produced gases within the AnMBR sludge suspension is also assumed to be minimal compared to sparger gas flowrates.
2.8 REFERENCES


[37] I. Takács, ACTIVATED SLUDGE MODELLING.
ABSTRACT

This chapter compares the filtration behaviour of three model foulants which are representative of the proteins, carbohydrates and lipids found in high solids and fatty industrial wastewaters. The model foulants include egg white powder (protein), α-cellulose (carbohydrate) and animal fat (lipids). Flux-step analyses were conducted with single foulants to establish differences in their fouling behaviour and with binary and ternary combinations of each foulant to identify combined effects on membrane fouling. Proteins and protein-containing mixtures had the highest fouling rates compared to single lipid and carbohydrate solutions. Compressible fouling cakes were only observed in pure protein and in protein-containing mixtures. Furthermore, the extent of compressibility (quantified by the pressure, $P'$, at which initial specific cake resistance is doubled) was directly related to protein content; with similar values of $P'$ in mixtures comprising similar protein percentages. In mixtures containing both protein and lipids a synergistic effect of the mixtures on fouling rates was observed; and was attributed to the formation of large protein-lipid complexes which due their hydrophobic nature are reactive and can form highly viscoelastic fouling cake layers. Lipid filtration irreversibly fouled the membrane suggesting a primarily pore fouling mechanism in lipids. However, in the presence of proteins a primarily cake fouling mechanism was observed, and can be attributed to protein adsorption of lipids. Hence, in fatty wastewaters, the higher fouling rates (and hence lower flux) resulting from the addition of a protein emulsifier may be worthwhile if it means protecting the membrane from irrecoverable pore fouling and extending membrane life. AnMBR sludge, which comprises a large proportion of insoluble protein (90%) and carbohydrate (9%) behaves in accordance with the fouling and compressibility behaviour shown by the protein and protein-carbohydrate mixtures.
3.1 INTRODUCTION

The potential of anaerobic membrane bioreactors (AnMBRs) for the treatment of high strength (high chemical oxygen demand and solids) industrial wastewaters has been comprehensively reviewed in literature [1-3]. AnMBRs are particularly attractive in the treatment of wastewaters with high lipids and solids as they combine high rate anaerobic treatment (HRAT) with membrane biomass retention that is independent of sludge settleability [4]. Food and animal processing industries typically generate waste streams high in lipids and solids; hence providing an opportunity for the use of AnMBRs in treatment of wastewaters from these industries. Existing research into industrial AnMBRs are generally focused on biological treatment capability, with an aim of achieving short hydraulic retention times (HRTs) to meet high rate treatment requirements. There is limited focus however on fouling in industrial AnMBRs treating complex and fatty wastewaters. The membrane fouling propensity of a wastewater directly affects operating membrane flux and hence HRT and organic loading rates (OLR). Therefore membrane flux is a key factor determining overall AnMBR treatment capacity. Operating flux is directly linked with overall MBR costs, and there exists a critical flux below which costs (per permeate volume) steeply rise [5]. The feasibility of AnMBRs for treatment of complex industrial wastewaters hence depends on minimising membrane fouling and achieving sustainable and sufficiently high, long term operating fluxes.

It is generally expected that the factors affecting membrane fouling in AnMBRs will be similar to those in aerobic/activated sludge membrane bioreactors [1]. These factors include, membrane characteristics (pore size, hydrophobicity etc.), reactor hydrodynamics, mixed liquor suspended solids (MLSS), temperature, pH, particle size distribution (PSD) and soluble microbial products (SMP) and extracellular polymeric substances (EPS) [3, 6]. Le Clech et al [7], in their review on membrane fouling in activated sludge MBRs suggest that in most cases MBR fouling is attributed to the interaction between the membrane and the biological suspension (through MLSS, SMP and EPS) rather than between membrane and substrate, and that any effect of substrate on fouling is indirect via its effect on biomass. This may well apply to most domestic applications; however, in HRAT of industrial wastewater with an accumulation of slowly degrading particulates, the effect of substrate on fouling propensity may be significant and complex and hence requires further investigation. Streams from industries such as food, wood and animal processing especially contain high concentrations of particulate proteins, lipids and carbohydrates/polysaccharides. These slowly degradable particulates can accumulate over long periods in MBR systems [8] and therefore will be expected to affect bulk filtration properties.
Globular proteins such as blood (serum albumin), egg albumins such as ovalbumin and milk derived proteins such as casein and whey proteins, are commonly used/applied/produced in the food, dairy and meat industry. Particulate carbohydrates such as lignin, cellulose, hemicellulose and lignocellulosic materials are common in wood and animal processing industries [9, 10]. High fats, oils and grease (collectively termed lipids in this chapter) are also characteristic in some food wastewaters and in abattoir rendering waste streams and can have typical lipid concentrations of around 3000 mg/L. Lipids in food and animal processing industries can exist in free and emulsified forms [11]. AnMBRs are generally ideal for the treatment of stable lipid emulsions, and although free lipids can be pre-treated via mechanical separation, the high methane potential of lipids provides a case for their retention in the AnMBR feed stream [4, 11-13]. Retention of free lipids however, can cause severe fouling problems [4], and any benefits related to methane yield and energy recovery will have to be weighed against lipid fouling propensity and hence achievable membrane flux.

In wastewater membrane bioreactor (MBR) research, representation of EPS and SMP by model proteins, such as casein and bovine serum albumin, and model polysaccharides such as starch, alginate, chitosan, xanthan gum, glucan and dextran, have offered insights into the differences in fouling behaviour of proteins and polysaccharides [14-20]. Fundamental analysis using model foulants provide a reference against which behaviour of actual MBR foulants can be compared [17]. In submerged MBRs, performance can be determined via comparison of experimentally determined critical flux. This follows the original critical flux hypothesis that a critical flux exists below which permeability decline over time does not occur, and above which fouling is observed [7, 21]. Critical flux is determined via flux-step experiments where flux is stepwise increased and the resulting transmembrane pressure is monitored for its stability. The flux at which TMP transitions from stable to unstable behaviour is identified as the critical flux [22].

There are limited fundamental investigations into the membrane fouling behaviour and mechanisms of insoluble protein, carbohydrate and lipids. Zhou et al (2015) [23] studied the fouling mechanisms of rendering wastewater (post dissolved air flotation) and found high cake resistance and compressibility of filter cakes that mostly comprised protein and lipids. However, this analysis was done in small-scale, dead-end and cross-flow configurations that are not representative of submerged MBR fouling; and the precise effects of protein, carbohydrate and lipid fractions on membrane fouling were not analysed. The fouling behaviour of fatty wastewaters has received limited attention [4, 24]. However, fundamental insight into the behaviour of lipids in combination with protein and carbohydrates is still required.
This study compares the behaviour (via flux-step analyses) of three model foulants which are representative of the insoluble proteins, carbohydrates and lipids found in high solids and fatty industrial wastewaters. These include egg white powder (protein), α-cellulose (carbohydrate) and animal fat (lipids). Flux-step analyses were conducted with single components to establish differences in their fouling behaviour and with binary and ternary combinations of each component to identify combined effects on membrane fouling. Additional flux-step analyses were conducted with mature sludge from a pilot scale AnMBR treating cattle slaughterhouse wastewater and comparisons are drawn between fouling behaviour of the model foulants and AnMBR sludge. Particle size analysis, contact angle of fouled membranes and cake resistance estimation offer additional insight into fouling mechanisms of the model foulants.

3.2 EXPERIMENTS

3.2.1 Design of MBR

**Lab-scale MBR**

Model foulant flux-step analyses were conducted in a 45 L rectangular tank (0.41 m length, 0.2 m width and 0.55 m wetted height) (Figure 3-1a). A submerged A4 flat sheet membrane (Aquatec-Maxcon Pty Ltd, Australia) was used for filtration, and was installed using a vertically mounted frame and sparger module. The frame encompasses the membrane entirely with a channel spacing of 5 mm on either side of the membrane to the frame wall. The bottom of the frame was arched outwards to capture majority of sparged gas bubbles and hence maximise shear on the membrane. The membrane was sparged with nitrogen gas at a sparge rate of 4 L/min. Lab-scale experiments were conducted at room temperature (approximately 25 ºC).

**Pilot scale MBR**

The pilot-scale AnMBR comprised a 200 L bioreactor (0.47 m diameter by 0.865 m wetted height) (Figure 3-1b) with two vertical mounted submerged A4 flat sheet membranes (Aquatec-Maxcon Pty Ltd, Australia). As in the lab-scale membrane frame, membrane channel spacing was 5 mm and membrane frame was also arched outward at the bottom to capture sparged gas. The pilot-scale sparger unit had the same surface area as the lab-scale unit. The membranes in pilot-scale were sparged with nitrogen gas at a rate of 3.5 L/min. Temperature in the AnMBR was measured using an RTD sensor (model SEM203 P, W&B Instrument Pty.), and was controlled at 32 ºC using heating tape. The effect of temperature on flux is assumed negligible, as differences in sludge
viscosity between 32°C and 25°C at dilute solids concentrations is minimal and close to water viscosity.

While there is expected to be hydrodynamic differences between the pilot scale AnMBR and the lab-scale tank, local hydrodynamics around the membrane are expected to be similar in both layouts due to the similar gas sparging rates and channel width between membrane sheet(s) and membrane frame wall.

![Figure 3-1: Schematics of submerged membrane bioreactor equipment used in flux-step experiments at (a) lab scale (45 L) and (b) pilot scale (200 L).](image)

3.2.2 Foulants

**Model foulants**

Egg white powder was obtained from an online supplements store (Bulk Nutrients, Australia); α-cellulose was sourced from Sigma Aldrich (C8002 Sigma) and animal fat (edible tallow) was bought from a local supermarket. Animal fat solutions were prepared by blending the required amount in a small amount of water for 60 s before adding this solution to the tank. Experimental design was based on a triangle matrix, similar to the three-substrate approach of Astals (2013) [25]. Each model foulant was tested individually and in combination in accordance with the matrix in Figure 3-2. A total solids (TS) concentration of 2000 mg/L was maintained in all experiments.
Pilot scale AnMBR sludge characteristics

The pilot-scale AnMBR treating slaughterhouse wastewater had a total solids concentration of 2200 mg/L, with combined insoluble protein, polysaccharides and fats, oil and grease (FOG) concentrations of 1487 mg/L. Protein, followed by carbohydrate, made up the significant fraction of total insoluble organic compounds, with protein/carbohydrate/lipid (P/C/L) proportions at approximately 90/9/1. Total protein was estimated using the BCA method (Sigma Catalog Nos. BCA1 and B9643) with bovine serum albumin (BSA) standard. Carbohydrate was estimated using the Anthrone method with D-glucose standard. Fats, oil and grease were measured using Standard Methods 5520. Total solids (TS) concentration was measured using Standard Methods 2540B.

Table 3-1 summarises the characteristics of substrate and wastewater used in the filtration experiments.
Table 3-1: Substrate and wastewater characteristics

<table>
<thead>
<tr>
<th>Model foulant description</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Model protein</td>
<td>Egg white (albumen) powder</td>
</tr>
<tr>
<td>Model carbohydrate</td>
<td>α-cellulose</td>
</tr>
<tr>
<td>Model lipid</td>
<td>Animal fats</td>
</tr>
<tr>
<td>Concentration maintained in each experiment</td>
<td>2000 mg/L</td>
</tr>
</tbody>
</table>

AnMBR sludge characteristics

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Insoluble protein</td>
<td>1334 mg/L</td>
</tr>
<tr>
<td>Insoluble polysaccharides</td>
<td>135 mg/L</td>
</tr>
<tr>
<td>Lipids</td>
<td>18 mg/L</td>
</tr>
<tr>
<td>Total solids concentration</td>
<td>2200 mg/L</td>
</tr>
</tbody>
</table>

3.2.3 Particle size

The presence of finer particles is important in membrane fouling as smaller particles are preferentially dragged towards the membrane [26], and hence can lead to compressible cakes with high resistance leading to lower critical membrane flux [27]. Additionally, the monitoring of mean particle sizes and distributions in each individual foulant solution and mixture will provide an indication of whether particles undergo aggregation when mixed with each other.

Particle size distributions of feed solutions were measured using a Malvern Mastersizer (laser diffraction method). Fully mixed samples of each foulant (individual and combined) solution were used; with five measurements per sample. Key parameters in particle size distributions include the 50th percentile point (d50), which represents the median diameter of the distribution; and moment means such as the De Brouckere mean (D[4,3]) and Sauter mean (D[3,2]). D[4,3] is the weighted-average volume diameter assuming spherical particles of the same volume as the measured particles and is sensitive to the presence of large particles. D[3,2] or surface area mean diameter assumes spherical particles with the same surface area as the measured particles and is sensitive to the presence of finer particles. D[3,2] is important in applications where particle surface area is important, and is often used in cake filtration studies (for example the Kozeny-Carman equation which defines pressure drop across a porous cake as a function of its porosity and Sauter mean).

3.2.4 Contact angle

Hydrophobic substances such as protein and lipids are more likely to attach on to hydrophobic membrane surfaces [28]. Foulant deposition can also change the hydrophobicity of the membrane surface; which will then govern flux and fouling rates [29]. Therefore measurement of membrane surface hydrophobicity before and after filtration will provide useful explanations for the
differences in fouling behaviour in the tested foulants. Contact angle is the angle formed by a water
droplet on the surface of interest, and was measured using an OCA 20 contact angle meter (sessile
drop method. A large contact angle indicates that wetting of the surface is unfavourable; i.e. the
surface is hydrophobic so the droplet will minimise contact with the surface.

The OCA 20 comprises a high resolution CCD (charge-coupled device) camera with a mechanical
dosing arm which drops precise volumes of water on the surface of interest. A volume of 12 µL was
dispensed at a rate of approximately 2-4 µL/s. An image of the water drop was captured
approximately 2 s after the drop was dispensed. Maintaining 2 s before snapping of the image
allowed the drop to properly form after being dispensed by the syringe. This also provided
consistency in contact angle estimations of all surfaces; particularly since drop shapes and sizes can
change within seconds of dispensing.

### 3.2.5 Flux-step experiments

Filtration or flux-step experiments were conducted in accordance with the protocol described by Le-
Clech et al (2003) [22]. In this protocol, flux is stepwise increased and the resulting transmembrane
pressure (TMP) is recorded. Fouling rate, dP/dt, was taken as the gradient of the TMP line at each
flux step. The behaviour of the dP/dt vs. flux curve was used to comment on the fouling propensity
of the substrate tested, with higher rate of increase in dP/dt (fouling rate) indicating greater fouling
propensity. The flux at which dP/dt exceeds 0.01 KPa/min was taken as the critical flux, in
accordance with the criteria proposed by Le-Clech et al (2003) [22]; although visual observations of
dP/dt behaviour were also considered when estimating critical flux.

In the lab scale experiments flux was incrementally increased in steps of 5 LMH, terminating at a
final flux of 60 LMH (or when permeate flow ceased indicating complete membrane fouling). Flux
steps were of 5 minute durations each. In pilot scale, flux was incrementally increased in steps of 4
LMH, terminating at a final flux of 17 LMH. Each filtration test was conducted in duplicate after
ensuring membranes in each test had the same initial clean water resistances.

A Watson Marlow peristaltic pump (model: 520 Du) was used to maintain and control flux during
the flux-step experiments. Pressure change was continuously monitored by means of a pressure
transducer (model Druck PTX 1400, GE) situated at the permeate stream. The permeate stream was
recirculated into the tanks to maintain the liquid level. Pressure transducer output signal (4-20 mA)
was logged via PLC onto a computer. Circulation in the lab and pilot scale tanks was achieved
using a Masterflex pump (model: 253G-200E-W1219) at flow rate of 12 L/min and 10 L/min, respectively, to avoid particle settling.

Membranes were systematically cleaned prior to each measurement. Cleaning protocol included rinsing in tap water, after which they were soaked in calcium hypochlorite (effective chlorine concentration of 0.3 wt%) and subsequently 0.2 wt% citric acid for 2 hours each. Lipid filtration tests were usually followed by physical cleaning in a warm solution (37 to 43°C) of 0.5 wt% sodium laureth sulphate before routine chemical cleaning. The membranes were rinsed again and stored in tap water until they were next required. Clean water permeability was measured subsequently to evaluate permeability recovery prior to the next trial. An arbitrary permeability recovery baseline of 95% was chosen, below which the membrane was discarded.

Experimental conditions are summarised in Table 3-2.

**Table 3-2: Summary of experimental conditions**

<table>
<thead>
<tr>
<th>Lab-scale tank operation</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Tank wet volume</td>
<td>45 L</td>
</tr>
<tr>
<td>Recirculation rate</td>
<td>12 L/min</td>
</tr>
<tr>
<td>Nitrogen gas sparge rate</td>
<td>4 L/min</td>
</tr>
<tr>
<td>Flux range</td>
<td>5 to 60 LMH</td>
</tr>
<tr>
<td>Flux step size</td>
<td>5 LMH</td>
</tr>
<tr>
<td>Flux step duration</td>
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</table>

<table>
<thead>
<tr>
<th>Pilot-scale tank operation</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Tank wet volume</td>
<td>200 L</td>
</tr>
<tr>
<td>Recirculation rate</td>
<td>10 L/min</td>
</tr>
<tr>
<td>Nitrogen gas sparge rate</td>
<td>3.5 L/min</td>
</tr>
<tr>
<td>Flux range</td>
<td>4 to 17 LMH</td>
</tr>
<tr>
<td>Flux step size</td>
<td>4 LMH</td>
</tr>
<tr>
<td>Flux step duration</td>
<td>15 min</td>
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<table>
<thead>
<tr>
<th>Membrane cleaning and clean water resistance</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Calcium hypochlorite (70% effective chlorine)</td>
<td>0.3 wt%</td>
</tr>
<tr>
<td>Citric acid</td>
<td>0.2 wt%</td>
</tr>
<tr>
<td>Duration of chemical cleaning</td>
<td>2 h each</td>
</tr>
<tr>
<td>Sodium laureth sulphate concentration in 43°C water (after lipid tests only)</td>
<td>0.5 wt%</td>
</tr>
<tr>
<td>Initial membrane resistance (clean water)</td>
<td>$5.78 \times 10^{10} \pm 3.97 \times 10^9 \text{m}^{-1}$</td>
</tr>
</tbody>
</table>
3.3 MODEL BASED ESTIMATION OF CAKE RESISTANCE PARAMETERS

Cake resistance to flow affects the critical flux at which an AnMBR can operate. In cake layers that are compressible, cake resistance increases with applied TMP; which hence reduces the critical flux, compared to that of a cake that is incompressible. Cake compressibility is typically high in ‘soft’ deformable sludge and in cakes with small particle sizes that can structurally rearrange themselves into smaller gaps when pressure is applied [30]. Cake resistance parameters such as specific cake resistance and cake compressibility are related via the empirical equation 3-1 [31, 32]:

\[ r_c = r_{c0} \left( 1 + \frac{P}{P'} \right) \]  

(3-1)

Where:
- \( r_c \) = specific cake resistance [m/kg]
- \( r_{c0} \) = initial specific cake resistance (at zero applied TMP) [m/kg]
- \( P \) = TMP [Pa]
- \( P' \) = TMP at which \( r_{c0} \) doubles [Pa]

In compressible cakes, \( P' \) is low; i.e. the specific cake resistance is doubled at a lower applied TMP. In incompressible cakes \( P' \) is high, hence \( r_c \approx r_{c0} \).

Specific cake resistance and compressibility are easily estimated in dead-end configurations, where the cake layer is preserved and hence cake mass is exactly known. However, in submerged configurations, the cake layer is lost as soon as applied pressure is removed; and hence direct in-situ measurements of specific cake resistance and compressibility are impossible. Numerically fitted measured response of TMP and flux has been used in literature to derive the values of specific cake resistance and compressibility in non dead-end configurations [33]. Here, a membrane fouling model, detailed in Chapter 4 of this thesis and in Boyle-Gotla et al (2014) [34] is used for estimation of cake resistance parameters. This model predicts cake accumulation on the membrane surface using a membrane shear profile obtained from a computational fluid dynamics (CFD) model of the lab scale AnMBR Predicted cake mass is then used to estimate total resistance and the subsequent dynamic TMP response to step-wise increase in flux. An overview of the model is outlined in the schematic in Figure 3-3. For full details of the model, please refer to Chapter 4.
Figure 3-3: Model overview. Estimated parameters (initial specific cake resistance and compressibility) are highlighted in red

The values of cake resistance parameters, $r_{c0}$ and $P'$ for each of the foulants tested in this study were obtained using parameter estimation while fitting modelled flux-pressure relationships against those obtained during flux-step experiments. Parameter estimation uses a non-linear least squares parameter estimation method (Levenberg-Marquardt algorithm) with TMP as fitted output and residual sum of squares ($J$) as objective function. Parameter uncertainty was estimated from a two-tailed t-test (95% confidence interval) on the parameter standard error, calculated from the objective-parameter Jacobian at the optimum (i.e., linear estimate) [35].

Membrane shear is estimated via 3D CFD modelling of the lab scale MBR. A two fluid (Eulerian-Eulerian) approach was used to simultaneously model liquid and gas phases. The liquid phase was assumed to follow Newtonian rheology, with a constant viscosity of 0.001Pa.s (water viscosity at ~20°C). Estimates of cake resistance and compressibility for pilot-scale AnMBR sludge (Chapter 4) are also presented in this chapter for comparison purposes.

Fluid turbulence was modelled using a $k-\varepsilon$ turbulence model, which is the most widely validated of the existing Reynolds Stress turbulence models [36, 37]. Default values of the empirical turbulence constants are used in the CFD analysis, in accordance with the standard values provided by Rodi [36]. Additional models include the use of a Grace drag model for gas-liquid momentum transfer,
Tomiyama lift force model and Sato’s model for turbulence enhancement in bubbly flows. Full details of the above CFD sub-models are provided in Chapter 2 of this thesis.

The lab-scale MBR domain was meshed using ANSYS meshing software. The mesh comprises a total approximately 3.6 million elements and is within quality recommendations (orthogonal quality >0.1, skewness <0.95). The solver was run in steady-state mode until monitored parameters (fluid velocity, maximum and average wall shear) reached stable values and the maximum residual target was $1 \times 10^{-3}$ was reached.

3.4 RESULTS

3.4.1 Particle size distributions

Table 3-3 summarises key values from particle size analysis of the model foulants (individual and in combination). Particle size distribution graphs are attached in Appendix Figure A-1.

Table 3-3: Mean (volume and surface) and median particle sizes, ($\mu$m)

<table>
<thead>
<tr>
<th></th>
<th>D[4,3]</th>
<th>D[3,2]</th>
<th>d50</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protein</td>
<td>23.5</td>
<td>5.1</td>
<td>11.3</td>
</tr>
<tr>
<td>Carbohydrate</td>
<td>89.7</td>
<td>22.2</td>
<td>50.3</td>
</tr>
<tr>
<td>Lipids</td>
<td>7.3</td>
<td>2.4</td>
<td>3.0</td>
</tr>
<tr>
<td>P/C (50/50)</td>
<td>133</td>
<td>28.2</td>
<td>77.7</td>
</tr>
<tr>
<td>P/C (10/90)</td>
<td>128.2</td>
<td>32.2</td>
<td>78.5</td>
</tr>
<tr>
<td>P/C (1/99)</td>
<td>127.7</td>
<td>32.3</td>
<td>78.0</td>
</tr>
<tr>
<td>P/L (50/50)</td>
<td>133.4</td>
<td>5.2</td>
<td>51.3</td>
</tr>
<tr>
<td>P/L (10/90)</td>
<td>178.3</td>
<td>8.6</td>
<td>118.8</td>
</tr>
<tr>
<td>C/L (50/50)</td>
<td>133.2</td>
<td>32.4</td>
<td>83.1</td>
</tr>
<tr>
<td>P/C/L</td>
<td>133.3</td>
<td>8.5</td>
<td>61.2</td>
</tr>
<tr>
<td>AnMBR sludge</td>
<td>68.8</td>
<td>27.5</td>
<td>50.6</td>
</tr>
</tbody>
</table>

Of the individual component solutions, lipids had the smallest volumetric median ($d_{50}$) of 3.0 $\mu$m followed by protein (11.3 $\mu$m) and carbohydrate (50.3 $\mu$m). Values for D[4,3], D[3,2] and $d_{50}$ were in the same order of magnitude, suggesting a significant proportion of fine lipid particles. Protein solutions were similarly skewed towards the smaller size range, with D[4,3], D[3,2] and $d_{50}$ having
the same order of magnitude. Carbohydrate had a bimodal distribution (Appendix Figure A-1a) with a D[4,3] and D[3,2] of 89.7 µm and 22.2 µm, respectively.

Protein-carbohydrate binary mixtures had higher d_{50} than their individual components. D[4,3] and D[3,2] in P/C mixtures were similar to those of pure C, suggesting that the mixture takes on the particle size properties of C. This is confirmed by the fact that all P/C mixtures had similar distributions; which also indicates an independent relationship between particle size and P/C ratios.

P/L mixtures had a higher d_{50} than individual components, with P/L (50/50) and P/L (10/90) having d_{50} values of 51.3 µm and 118.8 µm, respectively. The P/L particle size distributions covered a wide range, with P/L (50/50) showing greater polydispersivity than P/L (10/90) (Appendix Figure A-1c). This is reflected in the relatively large D[4,3] and small D[3,2] values of the P/L mixtures. Hence P/L shows evidence of particle aggregation; but also the maintenance of finer particle sizes. The D[3,2] in P/L mixtures are similar to pure P, suggesting that P contributes to the finer particle sizes.

The C/L (50/50) mixture had a d_{50} of 83 µm, higher than its individual components. The distribution and values of D[4,3] and D[3,2] in this mixture was similar to P/C mixtures and pure C mixtures; suggesting that C is the dominant contributor to the particle size distribution. The P/C/L combination had a wide distribution of particles, and it was difficult to discern a clear peak (Appendix Figure A-1e). Values of D[4,3] and D[3,2] were similar to P/L mixtures.

AnMBR sludge, comprising mostly protein (90%) and carbohydrate (9%) had particle size distributions approximately overlapping the pure P and P/C (50/50) mixture (Figure 3-4).
AnMBR sludge and P/C mixtures also had similar D[3,2] of approximately 28 µm. Unlike the P/L and P/C/L mixtures, AnMBR sludge had a unimodal distribution and relatively low D[4,3], with no evidence of particle aggregation. Hence it can be inferred that due to the low lipid content (1%) in AnMBR sludge, protein-lipid aggregation does not occur to the same extent as in the P/L and P/C/L mixtures.

3.4.2 Material hydrophobicity

Contact angle for new and fouled membranes are summarised in Table 3-4. Images of the sessile drop are attached in Appendix Figure A-2.

Table 3-4: New and fouled membrane hydrophobicity

<table>
<thead>
<tr>
<th>Foulant</th>
<th>Average contact angle (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>New (unused) membrane</td>
<td>38.0 ± 2.8</td>
</tr>
<tr>
<td>Protein (0.155 g)</td>
<td>81.2 ± 2.2</td>
</tr>
<tr>
<td>Protein (1.0 g)</td>
<td>82.6 ± 1.1</td>
</tr>
<tr>
<td>Carbohydrate</td>
<td>Not measurable</td>
</tr>
<tr>
<td>Lipids</td>
<td>102.9 ± 0.4</td>
</tr>
</tbody>
</table>

An unused flat sheet membrane (chlorinated polyethylene) is hydrophilic as indicated by the low contact angle of 38°. The contact angle increases in protein fouled membranes to approximately 80° in both 1.0 g and 0.155 g protein. Hence hydrophobicity is independent of cake mass. Lipid fouled membranes had the greatest hydrophobicity of all membranes (103°). Carbohydrate fouled membranes were also tested; but the dispensed drop was immediately flattened. Hence it can be concluded that carbohydrate cake is extremely hydrophilic.

3.4.3 Filtration behaviour

Figure 3-5 displays the change in transmembrane pressure with time (dP/dt) at each flux step during the filtration of individual feed components. Flux units of L/m²-h, or LMH, are used.
Figure 3-5: $dP/dt$ vs. flux for a) individual components; b) P/C mixtures; c) P/L mixtures; d) C/L mixtures; e) P/C/L mixtures f) AnMBR sludge vs individual substrate and P/C (50/50) and P/L (50/50) mixtures. Some $dP/dt$ lines are repeated in more than one graph for comparison purposes, and are represented by dotted lines.
Figure 3-5a displays the fouling rates of individual foulants, P, C and L. Protein had the highest fouling propensity, reaching a maximum dP/dt of 0.088 kPa/min at a flux of 20 LMH. Above 20 LMH, no further filtration was possible (due to complete fouling of the membrane) indicating this was the maximum achievable flux. dP/dt exceeded 0.01 kPa/min within the first flux step of 5 LMH, therefore the critical flux for proteins was below 5 LMH (dP/dt of 0.016 kPa/min).

Fouling rate (dP/dt) of lipids was lower than that of protein. A critical flux for lipids was identified at 25 LMH. Above 40 LMH, a stable dP/dt of 0.049 kPa/min was maintained indicating no further increase in fouling. Carbohydrates had the lowest overall fouling rate with a relatively flat profile. dP/dt exceeded 0.01 kPa/min at a flux of 40 LMH, but only slightly increased subsequently to a stable value of approximately 0.015 kPa/min. This relatively stable dP/dt indicates that critical flux of the carbohydrate suspension may exceed 40 LMH.

dP/dt of protein/carbohydrate (P/C) mixtures

Figure 3-5b displays the dP/dt profiles with varying compositions of proteins and carbohydrate (P/C of 50/50, 10/90 and 1/99, respectively). Of the mixtures, the worst fouling was observed in P/C of 50/50 followed by 10/90 and 1/99. The dP/dt curves of the mixtures lay between those of the individual components, i.e. as carbohydrate composition was increased, fouling behaviour tended towards that of pure carbohydrates and vice versa.

P/C (50/50) had a higher critical flux (10-15 LMH) than the pure protein solution. This mixture also had a higher maximum achievable flux (25 LMH) than the pure protein solution. The profile of P/C (10/90) lay approximately halfway between the profile of pure protein and carbohydrate. This mixture had a similar critical flux to P/C (50/50) (10-15 LMH), however it also had a slower overall increase in fouling rate and was therefore able to achieve a higher maximum flux (40 LMH), than P/C (50/50). P/C (1/99) had a flatter fouling profile closer to that of pure carbohydrate and was able to maintain filtration for the full range of experiment fluxes. This mixture had a higher critical flux (20-25 LMH) than P/C (50/50) and (10/90).

dP/dt of protein/lipid (P/L) mixtures:

Figure 3-5c displays the fouling rates of protein and lipid mixtures with P/L compositions of 50/50 and 10/90. In contrast to P/C mixtures, overall fouling rates for both P/L mixtures exceed the fouling rates of their individual components. P/L (50/50) had a higher fouling rate than P/L (10/90), indicating higher fouling at high P/L ratios. Critical flux was below the first flux step of 5 LMH for
both P/L mixtures. P/L (50/50) reached a maximum flux of 20 LMH while P/L (10/90) could reach a higher maximum flux of 35 LMH before failure.

\textbf{dP/dt of carbohydrate/lipid (C/L) mixture:}

Fouling rates for carbohydrate and lipids is shown in Figure 3-5d, with the dP/dt curve of the C/L (50/50) mixture lying between the dP/dt curves of its pure components. C/L (50/50) had a critical flux of 30 LMH.

\textbf{dP/dt of protein/carbohydrate/lipid (P/C/L) mixture}

Fouling rate with the ternary mixture comprising all three components in equal compositions (33/33/33) is shown in Figure 3-5e. Fouling profiles of individual components and their binary mixtures (50/50) are included in the figure for comparison. The dP/dt curve of the ternary mixture increased at a faster rate than the curves of the binary mixtures and individual components, with a critical flux less than 5 LMH and a maximum achievable flux of 20 LMH.

\textit{Model foulants vs. actual AnMBR sludge}

The dP/dt profile of pilot scale AnMBR sludge is shown in Figure 3-5f against the profiles of pure protein, carbohydrate and lipids and their 50/50 mixtures for comparison. AnMBR sludge fouling rate closely follows that of pure protein, and is between the profiles of P/L (50/50) and P/C (50/50).

Key filtration results are summarised in Table 3-5; with model foulants ranked from lowest to highest fouling propensity (prioritised by critical flux).
Table 3-5: Summary of key filtration results for the individual feed components and mixtures, arranged from lowest to highest fouling propensity

<table>
<thead>
<tr>
<th>Component</th>
<th>Critical flux (LMH)</th>
<th>Max. dP/dt (KPa/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbohydrate (C)</td>
<td>40</td>
<td>0.015</td>
</tr>
<tr>
<td>C/L (50/50)</td>
<td>30</td>
<td>0.03</td>
</tr>
<tr>
<td>P/C (1/99)</td>
<td>20 to 25</td>
<td>0.054</td>
</tr>
<tr>
<td>Lipid (L)</td>
<td>25</td>
<td>0.056</td>
</tr>
<tr>
<td>P/C (10/90)</td>
<td>10 to 15</td>
<td>0.064</td>
</tr>
<tr>
<td>P/C (50/50)</td>
<td>10 to 15</td>
<td>0.119</td>
</tr>
<tr>
<td>AnMBR sludge</td>
<td>8.6</td>
<td>0.084</td>
</tr>
<tr>
<td>Protein (P)</td>
<td>&lt;5</td>
<td>0.088</td>
</tr>
<tr>
<td>P/L (10/90)</td>
<td>&lt;5</td>
<td>0.105</td>
</tr>
<tr>
<td>P/L (50/50)</td>
<td>&lt;5</td>
<td>0.144</td>
</tr>
<tr>
<td>P/C/L</td>
<td>&lt;5</td>
<td>0.269</td>
</tr>
</tbody>
</table>

Irreversible fouling

Figure 3-6 displays membrane resistance before filtration; and after filtration (and physical and chemical cleaning) to identify membrane resistance recovery and irreversible fouling.

![Figure 3-6: Membrane resistance with clean water, before filtration and after filtration and physical and chemical cleaning.](image)
Apart from pure lipid and C/L mixture, membrane resistance in all experiments were almost fully recovered. While pure lipid filtration did not have the same fouling rates as pure protein filtration, it showed signs of irrecoverable fouling (i.e. membrane resistance was not recovered after chemical cleaning). The extent of irrecoverable fouling was not consistent in the four lipid tests, L(1) to L(4), as shown in Figure 3-6. This could be attributed to the varying rates of lipid agglomeration over the course of each experiment, which reduced the homogeneity of the lipid solution. The presence of protein appeared to protect the membrane from irrecoverable fouling as suggested by the complete recovery of membrane resistance in P/L and P/C/L mixtures.

### 3.4.4 Filter cake properties

Model estimated initial specific cake resistance, $r_{c0}$ and compressibility parameter, $P'$ are shown in Figures 3-7a and b, respectively.

AnMBR sludge and protein have similar cake properties; with respective $r_{c0}$ values of $5 \times 10^{13}$ m/kg and $4.0 \times 10^{13}$ m/kg and respective $P'$ values of $870 \pm 80$ Pa and $830 \pm 35$ Pa. Of the three individual foulants; protein had the highest $r_{c0}$, followed by lipids ($2.9 \times 10^{13}$ m/kg) and carbohydrates ($0.4 \times 10^{13}$ m/kg). Pure lipids and carbohydrates also formed incompressible cakes.
As protein content decreased from 50% to 1% in P/C mixtures, \( r_{c0} \) values also decreased from \( 11.5 \times 10^{13} \) to \( 1.1 \times 10^{13} \), respectively. The compressibility parameter, \( P' \) increased from 4706±260 Pa to 9994±12 as protein content in the P/C mixtures decreased from 50% to 10%, indicating a reduction in cake compressibility with protein content. Further decrease in protein content to 1% did not affect \( P' \) (9983±27 Pa). A similar trend was observed in P/L mixtures, with \( r_{c0} \) decreasing from \( 20 \times 10^{13} \) to \( 11 \times 10^{13} \) and \( P' \) increasing from 4972±869 Pa to 10051±554 Pa as protein content decreased from 50% to 10%.

The C/L mixture had a \( r_{c0} \) of \( 0.5 \times 10^{13} \) m/kg; which was in between the \( r_{c0} \) values of pure C and pure L. The C/L cake, like its individual constituents was incompressible. The P/C/L mixture had the highest \( r_{c0} \) of all tested foulants of \( 24 \times 10^{13} \). It had a \( P' \) of 5000±572 Pa; which is similar to \( P' \) of the P/C and P/L mixtures comprising 50% protein.

### 3.5 DISCUSSION

Experimental results confirmed the high fouling potential of protein, both as an individual component and as a mixture with carbohydrate and lipids. The higher fouling rates in protein-containing mixtures can be attributed to the greater hydrophobicity of protein which enhances particle aggregation; as well as the presence of finer particles (lower values of \( D[3,2] \)) in all protein-containing mixtures, which increase cake compressibility and resistance. However, fouling propensity cannot be attributed to particle size and hydrophobicity alone. In P/C mixtures, the \( D[3,2] \) particle size did not change; however the fouling propensity decreased as protein content decreased. A similar behaviour was observed in P/L mixtures. Lipid fouled membranes were also highly hydrophobic and pure lipid solutions also had particles in the smaller size range. However; the fouling rates in lipid solutions were not as high as in protein and did not produce compressible cakes. Lipids in the presence of protein, however; had the highest fouling rates of all binary mixtures. Hence fouling propensity appears to primarily be affected by the presence of protein; with hydrophobicity and particle size having secondary effects.

Compressibility is a key factor in the rate of TMP increase; with compressible cakes only observed in pure protein and in protein-containing mixtures. Furthermore, the extent of compressibility (value of \( P' \)) was directly related to protein content; with mixtures containing a similar proportion of protein having similar values of \( P' \). The lower initial specific cake resistance in pure proteins, compared with protein mixtures may be attributed to its higher surface charge which causes greater electrostatic repulsion within the cake layer creating a porous cake layer, with lower initial cake
resistance [38]. However, due to its high compressibility, specific cake resistance in protein cakes rapidly increase with applied TMP.

The effect of protein on fouling could additionally be due to its high chemical reactivity. The general mechanisms of protein fouling include initial physical adsorption onto the membrane surface followed by further protein build-up due to protein aggregation and hydrophobic interactions with membrane surfaces [39]. Protein aggregation primarily occurs due to the interaction of free thiol groups (carbon bound sulfhydryl) on protein molecules forming intermolecular disulphide bonds [40, 41]. Proteins with a large number of free thiol groups, like ovalbumin (egg white protein) used in this study, tend to form aggregates more easily and have the greatest rates of filtration flux decline [42-44]. The high reactivity of egg white protein hence explains the high fouling rates seen in the pure protein and protein mixtures.

Fouling in pure carbohydrate solution was minimal. This is consistent with findings from other filtration studies on particulate carbohydrates such as high viscosity hemicellulose [45] and in lignin extraction from black liquor- a high pH stream produced in the kraft pulping process [46, 47]. Carbohydrate in combination with protein, however, resulted in increased fouling rates (relative to pure carbohydrate). Protein-carbohydrate fouling rates were between that of pure carbohydrate and pure protein. However, the net effect of the mixture on fouling is not antagonistic, as even at low P/C of 1/99, fouling rates were higher than in pure carbohydrate suggesting a dominance of protein in the fouling behaviour of protein-carbohydrate mixtures. P/C fouling rates increased with protein content; confirming the importance of protein in P/C fouling. P/C mixtures had a higher $r_{c0}$ than pure C; which increased with increasing protein content. Unlike pure C solutions, P/C mixtures formed compressible cakes; with compressibility increasing at high protein content. In literature, P/C ratio has been identified as a key fouling indicator, with greater rates of fouling observed at higher P/C ratios [14, 16]. Denser cake layers with higher resistance to flow were also documented in protein and carbohydrate mixtures compared with single carbohydrate solutions [15, 17, 20, 48].

Protein-lipid combinations showed different fouling behaviour to protein-carbohydrate mixtures. While $dP/dt$ profiles of P/C mixtures lay between pure protein and pure carbohydrate profiles, the fouling rates of P/L mixtures were higher than pure protein and pure lipids. This suggests complex interactions between protein and lipids. Protein powders have the ability to entrap oil on their surfaces or in their hydrophobic cores. Proteins (mostly globular proteins from milk and eggs) are commonly used in the food industry to stabilise oil-in-water emulsions. In aqueous solutions, globular proteins such as albumins have the ability to unfold at the oil interface and expose their hydrophobic core to facilitate adsorption onto the oil droplet surface [49]. The net result is the
formation of a protein film on the oil droplet with its hydrophobic core oriented towards the oil phase and its outer hydrophilic surface oriented towards the water phase, therefore reducing oil agglomeration and increasing emulsion stability [49, 50]. The hydrophobic core contains reactive amino acids that are able to form hydrophobic and disulphide bonds with neighbouring proteins hence enhancing protein aggregation and forming a highly viscoelastic network [51-53]. Formation of reactive protein-lipid complexes is hence an explanation for the higher membrane fouling rates for P/L mixtures compared to pure P and pure L solutions. The presence of large particles, as indicated by the high D[4,3] in the P/L mixtures confirms increased particle aggregation.

Foaming was observed during the filtration of pure protein, P/C (50/50) and P/C (10/90). Foaming was especially enhanced when the sparger was operational, and hence can be attributed to physical denaturation of egg white during the sparging process. Physical denaturation of proteins, which can occur in high shear scenarios such as in high crossflow velocities, mixing and sparging, can enhance protein aggregation and hence membrane fouling [41, 54, 55]. Globular proteins such as BSA and other albumins found in milk and egg white have coiled spherical structures with an outer hydrophilic surface and an inner hydrophobic core. High shear near the membrane surface may expose the central hydrophobic region of proteins encouraging particle aggregation as well as adherence of these aggregates on a hydrophobic (fouled) membrane surface [55]. Denatured proteins can have greater fouling capacities than intact proteins due to exposure of hydrophobic groups [50] and may have also contributed to the high fouling rates observed in pure protein and P/C (50/50; 10/90). Little to no foaming was observed during P/L filtration; perhaps due to the adsorption of protein on lipid particles which stabilised the solution against physical denaturation.

Lipid fouling rates were lower than protein fouling rates, however, initial membrane resistance could not be recovered even after physical and chemical cleaning. Irreversible (removed by chemical cleaning) and irrecoverable fouling is generally caused by pore plugging and surface adsorption [56]. Literature on membrane filtration of fatty wastewaters such as, snacks factory wastewater [4], supermarket wastewater [57], household appliance factory wastewater containing emulsified mineral oil [58] and bilge water [59], found that the primary mechanism of lipid (including fats, oils and grease or FOG) fouling is adsorption of oil droplets within membrane pores which can lead to irreversible and irrecoverable membrane fouling. Therefore, lipid pore fouling and associated irreversible and irrecoverable membrane fouling will have a long term impact on fouling rates; which are not seen in the short term fouling rate measurements in this chapter. The fatty acid composition of a wastewater can also affect its fouling propensity. Oleic acid is the most dominant fatty acid in animal fats (beef tallow and lard) and this is significant as fatty mixtures containing oleic acid have the worst fouling propensity [28].
The fouling mechanism of lipids changes in the presence of protein from primarily pore fouling to primarily cake fouling, as indicated by the complete recovery of membrane resistance after P/L filtration. The change in fouling mechanism from pore fouling to cake formation, can be attributed to the formation of protein-lipid aggregates. A similar behaviour was documented in a study by Mueller et al (1997) [60] where suspended solids in an oily wastewater encouraged the formation of a secondary dynamic membrane (cake layer) and protected the underlying membrane from irreversible fouling. Hence the higher fouling rates (and hence lower flux) with the addition of a protein emulsifier in fatty wastewaters may be worthwhile if it means protecting the membrane from pore fouling.

The C/L mixture had a fouling rate in between that of pure C and pure L, with a slightly higher fouling capacity than pure C alone. α-cellulose powder is an inert carbohydrate which will not interact with lipids in the same way as protein. Hence, unlike the P/L mixture, membrane resistance in the C/L mixture was not recovered. This further confirms pore fouling behaviour of lipids, and the ability of protein to adsorb lipids and prevent pore fouling.

The P/C/L mixture had the highest fouling rates of all solutions tested in this study. This behaviour is attributed to interactions between the protein-oil-water emulsion and carbohydrate powder. The particle size distribution in the P/C/L mixture appeared to be a superposition of P/L and pure C size distributions. It is hence postulated that similar synergistic protein and lipid interactions take place in P/C/L mixture as in the P/L mixtures; with carbohydrate powder trapped in the viscoelastic P/L network, accelerating cake formation and hence fouling rates. In food applications, polysaccharides are used to increase stability of the protein-oil interface. Protein is initially added as the emulsifier followed by a polysaccharide which then interacts with the adsorbed protein forming a bilayer. This bilayer has increased charge and thickness therefore increasing stability against oil droplet agglomeration [49]. Polysaccharides are often added to oil-water food emulsions to enhance viscosity of the aqueous phase which imparts desirable textural attributes and stabilises the droplets against creaming [61]. Therefore increased fouling rates in the P/C/L mixture can be attributed to enhanced particle aggregation and viscosity. Like P/L, the P/C/L mixture also showed fully recoverable membrane resistance after chemical cleaning, confirming the surfactant and lipid binding effect of proteins.

Hydrophobic surfaces tend to foul at a faster rate; with surface modification to increase membrane hydrophilicity widely documented to improve filterability and limit fouling [62-67]. However, contact angle measurements suggest that original membrane surface hydrophilicity was reduced in the presence of a protein and lipid fouling layer indicating that once fouled, the membrane
characteristics take on those of the fouling layer. Hence membrane flux will become a factor of the fouling layer hydrophobicity rather than of the initial hydrophilic membrane after an initial filtration period and it is reasonable to suggest that in AnMBRs treating high concentration of hydrophobic particulates; hydrophilicisation during membrane fabrication will have little impact on fouling control.

Insoluble organic fraction in AnMBR sludge mostly comprises protein (90%) and carbohydrate (9%) and has fouling rates between pure protein and P/C (50/50). The remaining 1% lipid does not appear to have a significant effect on fouling rate. It is postulated that due to the low lipid content, protein-lipid synergistic fouling does not occur to the same extent as in the P/L and P/C/L mixtures; which is confirmed by the lack of larger particles in AnMBR sludge. The high protein content in AnMBR sludge gives it a similar cake compressibility (P'), to pure protein.

3.6 CONCLUSIONS

Of the three individual substrates studied here; protein had the highest fouling propensity. Carbohydrates and lipids had lower fouling rates individually than when mixed with protein also suggesting an important effect of protein on membrane fouling propensity. The higher fouling rates in protein mixtures can be attributed to greater hydrophobicity in protein which enhances particle aggregation; as well as the presence of finer particles (low D[3,2]) in all protein mixtures which increase cake compressibility and resistance. Compressible cakes were only observed in pure protein and in protein-containing mixtures. Furthermore, extent of compressibility was directly related to protein content. Protein-carbohydrate mixtures were dominated by the protein component, with protein-lipid mixtures fouled to a greater extent than pure proteins or pure lipids, indicating synergy. Irreversible fouling occurred mainly with pure lipids, with the presence of proteins mitigating irreversible fouling (at the expense of increased cake fouling). Hydrophobicity of the membrane surface is controlled by the fouling layer; and hence membrane flux will become a factor of the hydrophobicity of the fouled surface rather than of the initial membrane and suggests that surface modification of the membrane during membrane fabrication will have little impact on fouling control in particulate-laden wastewaters.
REFERENCES


4. DYNAMIC MULTIDIMENSIONAL MODELLING OF SUBMERGED MEMBRANE BIOREACTOR FOULING

ABSTRACT

Existing membrane fouling models are limited to simple hydraulic profiles which is a limitation particularly for planar membranes and for submerged membrane bioreactors where overall tank design can have a major impact on reactor hydrodynamics and hence membrane shear. This chapter presents a model that allows for a distributed shear profile, with dynamic linking of flux and transmembrane pressure (TMP). Shear profile is calculated using a multiphase computational fluid dynamic approach, and is applied to a distributed parameter model to simulate membrane fouling profile and flux distribution. This allows for simulation of complex flux-step experiments, or situations where non-uniform shear is present. The model was applied to filtration experiments conducted in a pilot-scale anaerobic membrane bioreactor treating slaughterhouse wastewater comprising 1950±250 mg/L total solids and was able to effectively fit experiments under dynamic critical flux conditions. Cake compressibility was a key parameter, and was estimated at 870±80 Pa. In non-uniform gas sparging, critical flux decreased from 12 LMH to 8.5 LMH. This emphasises the importance of local flow conditions on membrane fouling behaviour and that performance can depend heavily on reactor configuration and hydraulics.

This chapter was redrafted after
4.1 INTRODUCTION

Submerged membrane bioreactors (SMBR) are an established alternative to conventional clarifier based wastewater treatment technology with a number of advantages [1]. These include the removal of a separate clarification step and hence reduced footprint, better control over solids inventory and solids retention time, a more concentrated biomass and hence a higher space loading rate, and improved effluent quality due to elimination of solids through the membrane. Disadvantages are largely energy consumption related to membrane fouling management and permeate collection. The high cost of membranes and their replacement; which can be 50% of total capital expenditure [1], as well as energetic and chemical costs of fouling control and cleaning, provides strong motivation to predict and manage membrane fouling effectively.

Membrane fouling is the accumulation of largely insoluble material on or within the membrane [2], reducing membrane flux at a given transmembrane pressure (TMP) or conversely, increasing TMP at a given flux. It is possible to control membrane fouling during operation by gas sparging to create shear across the membrane and thus remove foulant. Physical and chemical cleaning operations are also used to periodically clean membranes, but require an interruption to process operation and are therefore less desired.

Membrane life can be enhanced by operating the SMBR below its ‘critical flux’; or the flux below which resistance to flow is governed by inherent membrane, rather than cake resistance [2]. Critical flux is generally determined in flux-step experiments, where membrane flux is step-wise increased and the corresponding TMP continuously measured [1, 3]. Critical flux is assessed as the flux at which the fouling rate (or rate of TMP increase, dP/dt) increases substantially above the baseline [4]. Overall resistance to flux is dominated by membrane resistance at subcritical flux and controlled by the resistance of the fouling layer at supercritical flux. Therefore, operation of an MBR below its critical flux is inherently more stable, with minimal periodic cake removal required (e.g. through backflushing). Critical flux is influenced by membrane shear, membrane properties, solids properties and membrane configuration [2]. It is therefore important to predict membrane and cake formation behaviour under subcritical, supercritical, and in transition fluxes.

Membrane fouling in SMBRs is most commonly assessed via experimental analysis [5]. This faces a number of challenges:
(1) The outcomes of a flux-step analysis are generally only applicable to MBR setups and process conditions similar to those tested in the experiment. In particular, lab-scale studies have limited applicability to full-scale applications due to different hydrodynamics [5].

(2) Hydrodynamics around the membrane creates surface shear on the membrane responsible for fouling control. Techniques for hydrodynamic characterisation include the use of dye particles [4, 6, 7], flow velocimetry [4, 8] and particle image velocimetry [9, 10]. These techniques however, cannot easily be implemented in large scale systems that have large volumes, variable flows and opaque fluids. They are also intrusive to flow and often restricted by the lack of space in reactor systems [11].

(3) Experimental analysis is generally limited to studying the impact of a small number of process variables at a given time and therefore may fail to account for the combined effects or interactions of the many process factors that affect membrane fouling [5]. These include; sludge properties such as solids concentration, stickiness, floc size, compressibility of cake layer and viscosity; and process parameters such as membrane flux, applied TMP and gas sparging intensity; which can vary between systems and over time.

Therefore, model based analysis is an important tool for fouling characterisation and effective fouling control, which allows for determination of platform independent fouling characteristics and a better fundamental understanding of controlling mechanisms.

Deterministic modelling of membrane fouling is at a comparatively early stage compared to general wastewater process and hydrodynamic modelling, partly due to its complexity. The default model approach is the use of lumped (non-distributed) parameter models which sufficiently characterise simple fouling behaviour [12-14]. These models however, are inapplicable in membranes with non-uniform shear and cake distributions and provide limited insight into transitional behaviour from satisfactory to unsatisfactory performance. A fractal permeation model was proposed by Meng et al (2005) [15] which estimates cake layer permeability by using fractal theory to characterise its microstructure. This model however has limited use as a predictive model as it is unable to estimate the impact of changing operational parameters and reactor conditions on cake resistance. Li and Wang (2006) [16] developed a semi-analytic, sectional resistance-in-series (RIS) fouling model which can be used to simulate dynamic sludge film formation on the membrane and its effect on TMP. This model considers net cake accumulation on the membrane to be dependent on the balance between accumulation due to membrane flux, which encourages solids deposition, and detachment due to shearing. This model can predict flux, cake thickness, for a given transmembrane pressure and bulk liquid shear, on the basis of parameters such as membrane resistance, solids stickiness and
compressibility. Further expansions and modifications to the model have since been made: Wu et al (2012) [17] included the effect of variable particle size solids, Zarragoitia-Gonzalez et al (2008) [18] integrated the model with the Activated Sludge Model No.1 (ASM1) to predict the impact of biologically produced soluble and insoluble products on membrane fouling and Mannina et al (2011) [19] further included deep bed filtration theory to predict chemical oxygen demand (COD) removal by the accumulated cake layer.

The sectional RIS fouling model and its extensions still have a number of limitations:
(a) The model geometry is 1-D and hence cannot assess the impact of variable shear and non-uniform fouling across the membrane surface
(b) Maximum shear intensity is correlated with aeration intensity via an empirical laminar correlation, and is therefore inaccurate for the prediction of shear in generally turbulent conditions. Shear profile along membrane length is also approximated by a sine function rather than a deterministic shear profile.
(c) The flux-pressure interaction is solved by iteration of the differential equation solution at a defined time point. This does not allow dynamic analysis of the flux-pressure interaction.

In particular, interaction of these limitations does not allow for detailed analysis of the interaction of fouling and flux distribution across a broader permeable domain.

In this chapter, a new approach to fouling modelling for a two dimensional membrane domain is presented. Membrane shear is calculated by a three dimensional, multiphase computational fluid dynamics (CFD) model. Pressure and flux are dynamically linked to resolve flux distribution across the domain and enable overall flux to be fixed.

4.2 MODEL OVERVIEW

Figure 4-1 provides an overview of the modelling approach used in this chapter. A three dimensional and two phase (gas-liquid) CFD simulation of the anaerobic MBR (AnMBR) configuration was used to estimate shear distribution on the membrane surface. This was then used with the fouling model of Li and Wang (2006) [16] to predict dynamic cake formation on the membrane. During a simulation, pressure is dynamically controlled using an integral controller such that the overall flux matches desired flux. Flux can be manipulated dynamically, which enables this model to simulate flux-step experiments for the prediction of critical flux for various operating parameters or normal subcritical operation.
Figure 4-1: Overview of the integrated CFD and fouling model developed in this study

4.2.1 CFD model prediction of membrane shear

A two fluid (Eulerian-Eulerian) approach was used to simultaneously model liquid and gas phases in the pilot scale AnMBR used for model development. Pilot scale AnMBR sludge contained a total solids (TS) concentration of 2 g/L, which in accordance with Equation 2-55 will have a viscosity of approximately 0.001 Pa.s at 34°C.

Fluid turbulence was modelled using a $k-\varepsilon$ turbulence model, which is the most widely validated of the existing Reynolds Stress turbulence models [20, 21]. Default values of the empirical turbulence constants are used in the CFD analysis, in accordance with the standard values provided by Rodi [20]. Additional models include the use of a Grace drag model for gas-liquid momentum transfer, Tomiyama lift force model and Sato’s model for turbulence enhancement in bubbly flows. Full CFD submodel details are provided in Chapter 2 of this thesis.

The AnMBR domain was meshed using ANSYS meshing software. The mesh comprises a total approximately 2.7 million elements and is within quality recommendations (orthogonal quality
>0.1, skewness <0.95). The solver was run in transient mode with timesteps of 0.01 s for a total duration of 120 s or until the model reached steady state; i.e. no variation in monitored parameters (fluid velocity, maximum and average wall shear). The maximum residual target was $1 \times 10^{-3}$.

4.2.2 **Fouling model**

**Sectional approach**

The membrane experiences non-uniform shear along its length and width, therefore resulting in uneven cake accumulation and flux across the membrane surface. To assess the space-variable shear, the membrane surface was discretised into a grid (finite element approach) comprising $34 \times 23$ or 782 sections.

**Cake accumulation and corresponding increase in TMP**

Cake accumulation rate, $(dM_c/dt)$, over each membrane section $(i,j)$, is described by a conservation equation, with the major mass transfer components of convective flux due to permeate movement through the membrane (positive) and loss due to transverse liquid shear (negative) related to gas sparging and cross-flow [2]. This conservation equation is described in Equation 4-2.

$$\left( \frac{dM_c}{dt} \right)_{ij} = \frac{24|TS|i^2}{24j+Ki'y} - \frac{\beta(1-\alpha)\gamma M_c^2}{\gamma/N_j\theta_f+M_c}$$

Accumulation = Attachment - Detachment

(4-2)

Solids attachment rate in Equation 4-2 describes convective movement of solids towards the membrane, which increases with membrane flux, $J$, and sludge solids concentration, $[TS]$ and decreases with transverse lift (lift coefficient, $K_l$) and membrane shear intensity, $\gamma$. The lift coefficient $K_l$, is a function of the particle drag coefficient, $C_D$ and particle size, $d_p$ ($K_l = C_d \times d_p$).

Solids detachment rate in Equation 4-2 is a function of sludge stickiness, $\alpha$, in the numerator and sludge compression, $\gamma'$, and filtration time, $\theta_f$, in the denominator. Therefore, at a given shear intensity, $\gamma$, a stickier cake layer (described by a higher stickiness coefficient, $\alpha$) is more difficult to detach. Higher sludge compression and longer filtration times also reduce sludge detachment rate.

The accumulated cake layer provides an additional resistance to flow at each membrane section, $R_{c,ij}$, which is calculated by Equation 4-3.
Resistance for each membrane section is calculated using the resistance-in-series approach, which describes overall resistance, $R_{ij}$, as the sum of inherent membrane resistance, $R_m$ and cake layer resistance, $R_{c,ij}$, and is given by equation 4-4.

$$R_{ij} = R_m + R_{c,ij} \quad (4-4)$$

$R_m$ is the primary contributor to total resistance at low flux when foulant accumulation is minimal to non-existent, and the flux versus TMP relationship is linear. At high flux, foulant build-up on the membrane increases total resistance and therefore the flux versus TMP relationship deviates from linearity. The transition point is at the critical flux. The strong form of the critical flux definition states that membrane resistance is the resistance to pure water, while the weak form defines membrane resistance as the total resistance at subcritical flux [2]. The weak form is applied here. Since this chapter mainly focuses on the impact of shear due to gas sparging on fouling or cake layer development, pore fouling is neglected in the model development. The contribution of pore fouling resistance to total resistance is also assumed negligible compared to that of cake fouling resistance [16].

In constant flux conditions, the increase in total overall resistance due to cake accumulation leads to an increase in TMP. Darcy’s law is used to describe the flux-pressure relationship, $J = \frac{P}{\mu R}$. Therefore, for a membrane section, individual flux, $J_{ij}$, and individual resistance, $R_{ij}$, are related to TMP by equation 4-5.

$$P = \mu J_{ij} R_{ij} \quad (4-5)$$

Dynamic flux-pressure interaction and the prediction of critical flux

During a flux-step experiment, dynamic increase in cake accumulation leads to a dynamic increase in TMP to maintain a period of constant flux during a flux-step. A virtual control loop is used to model this dynamic response, where the TMP is adjusted to minimise the error (or the difference between flux setpoint and actual flux) through a proportional-integral controller. This flux-pressure feedback loop enables the fixing of overall flux and the simulation of dynamic pressure increase despite flux distribution across the whole surface. The effect on TMP with stepwise increase in flux and the prediction of critical flux is hence made possible.
Model variables and parameters

\( R_m \) can be estimated by linear regression of \( \frac{dP}{dt} \) vs. flux below subcritical flux; and is therefore estimated as the total resistance at sub-critical flux.

Li and Wang used a constant \( r_c \) in their model. However, sludge cake is highly compressible [18, 22, 23] and its properties will vary with time during filtration in a constant flux scenario due to the increase in applied TMP. Therefore, \( r_c \) for a compressible material can be described by the empirical Equation 4-6 [24].

\[
r_c = r_{c0} \left( 1 + \frac{P}{P'} \right)^c
\]  

(4-6)

\( r_{c0} \) is the value of specific cake resistance at zero pressure and is assumed to vary between typical values of \( 3 \times 10^{12} \) m/kg to \( 3 \times 10^{14} \) m/kg for averagely dewaterable sludge [25]. A midrange value of \( 5 \times 10^{13} \) m/kg was adopted in this study. The compressibility coefficient, \( c \), can vary from 0 (non-compressible) to 1 (highly compressible). Activated sludge at 35°C was found to have a compressibility coefficient of approximately 1 [23], and this value was therefore adopted in this chapter. The compressibility parameter, \( P' \), is the TMP at which \( r_c \) is double its initial value, \( r_{c0} \). The value of \( P' \) is sludge specific as sludge cake compressibility can vary with floc size, sludge type, sludge conditioning, operating temperature and pH [13, 22, 26, 27]. Therefore, \( P' \) was estimated via parameter estimation during model calibration using a non-linear least squares parameter estimation method (Levenberg-Marquardt algorithm) with TMP as fitted output and residual sum of squares (J) as objective function. Parameter uncertainty was estimated from a two-tailed t-test (95% confidence interval) on the parameter standard error calculated from the objective-parameter Jacobian at the optimum (i.e., linear estimate) [28].

Flow near the sparger and membrane module is expected to be fully turbulent (Newton regime), where particle drag coefficient, \( C_D \), is approximately equal to 0.44 [29].

A typical value of the stickiness coefficient, \( \alpha \), of 0.5 was considered in this study. The remaining model parameters, erosion rate coefficient, \( \beta \), and compression coefficient, \( \gamma' \); (which is the actual rate of sludge compression measured as rate of change of solids concentration in cake layer and not to be confused with compressibility coefficient, \( c \)); were adopted from Li and Wang [16].

Fouling model variables and parameters are summarised in Table 4-1.
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>i,j</td>
<td>Section number</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$M_c$</td>
<td>Accumulated cake mass</td>
<td>Eq. 2</td>
<td>g/m²</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Shear rate</td>
<td></td>
<td>s⁻¹</td>
</tr>
<tr>
<td>TS</td>
<td>Total solids concentration</td>
<td>2</td>
<td>kg/m³</td>
</tr>
<tr>
<td>J</td>
<td>Membrane flux</td>
<td></td>
<td>L/m² h or LMH</td>
</tr>
<tr>
<td>$t$</td>
<td>Filtration time</td>
<td></td>
<td>mins</td>
</tr>
<tr>
<td>$V_f$</td>
<td>Volume of filtrate</td>
<td>$J \times t$</td>
<td>m³</td>
</tr>
<tr>
<td>$R$</td>
<td>Overall resistance</td>
<td>Eq. 4</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>$R_m$</td>
<td>Membrane resistance</td>
<td>$1 \times 10^{11}$</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>$R_c$</td>
<td>Cake resistance</td>
<td>Eq. 3</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>$r_c$</td>
<td>Specific cake resistance</td>
<td>Eq. 6</td>
<td>m/kg</td>
</tr>
<tr>
<td>$r_{c0}$</td>
<td>Initial specific cake resistance</td>
<td>$5 \times 10^{13}$</td>
<td>m/kg</td>
</tr>
<tr>
<td>c</td>
<td>Compressibility coefficient</td>
<td>1</td>
<td>[ ]</td>
</tr>
<tr>
<td>$P$</td>
<td>Transmembrane pressure</td>
<td>Eq. 5</td>
<td>Pa</td>
</tr>
<tr>
<td>$P'$</td>
<td>Pressure at which $r_c = 2 \times r_{c0}$</td>
<td>$870 \pm 80$</td>
<td>Pa</td>
</tr>
<tr>
<td>$d_p$</td>
<td>Particle diameter</td>
<td>60</td>
<td>µm</td>
</tr>
<tr>
<td>$C_D$</td>
<td>Drag coefficient</td>
<td>0.44</td>
<td>[ ]</td>
</tr>
<tr>
<td>$K_l$</td>
<td>Lift coefficient</td>
<td>$C_D \times d_p$</td>
<td>m</td>
</tr>
<tr>
<td>$\beta$</td>
<td>Erosion rate coefficient of sludge cake</td>
<td>$3.5 \times 10^{-4}$</td>
<td>[ ]</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Stickiness coefficient</td>
<td>0.5</td>
<td>[ ]</td>
</tr>
<tr>
<td>$\gamma'$</td>
<td>Compression coefficient</td>
<td>$2.5 \times 10^{-5}$</td>
<td>kg/(m³·s)</td>
</tr>
<tr>
<td>$n_{i,j}$</td>
<td>Number of membrane sections</td>
<td>782</td>
<td></td>
</tr>
<tr>
<td>$\mu$</td>
<td>Permeate viscosity</td>
<td>0.001</td>
<td>Pa·s</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>34</td>
<td>°C</td>
</tr>
<tr>
<td>$\theta_f$</td>
<td>Filtration period in a cycle</td>
<td>15</td>
<td>mins</td>
</tr>
</tbody>
</table>
4.3 EXPERIMENTS

The model developed here is calibrated and validated against two batch filtration experiments on a pilot scale AnMBR with fully digested slaughterhouse wastewater. The AnMBR comprised a 200 L bioreactor (0.47 m diameter by 0.865 m wetted height) (Figure 4-2) with two vertical mounted submerged A4 Kubota flat sheet membranes (Aquatec-Maxcon Pty Ltd, Australia). These were installed in an acrylic frame fitted with a gas sparger unit located 20 mm from the bottom of the membranes. Constant flux was maintained using a peristaltic pump on the permeate stream. Pressure change was continuously monitored by means of a pressure transducer (model Druck PTX 1400, GE) situated at the permeate stream. The permeate stream was recirculated into the bioreactor to maintain the liquid level within the reactor. Furthermore, temperature was controlled via an RTD sensor (model SEM203 P, W&B Instrument Pty.), and was maintained at the operating temperatures of the AnMBR (34 °C). Both pressure transducer and temperature output signal (4-20 mA) were logged via PLC onto a computer. Circulation at the bottom of membrane bioreactor was achieved using a Masterflex pump (model: 253G-200E-W1219) at flow rate of 613 L/h to avoid particle settling.

Figure 4-2: Experimental setup of the pilot scale FS and HF AnMBR

Membranes were systematically cleaned prior to each measurement. Cleaning protocol included physical cleaning with tap water and a soft sponge, after which they were soaked in 0.5 wt% sodium hypochlorite and subsequently 0.2 wt% citric acid for 2 hours each. The membranes were
rinsed again and stored in tap water until they were next required. Clean water permeability was measured subsequently to evaluate permeability recovery prior to the next trial.

Experiment 1 comprised a substrate (high protein slaughterhouse wastewater) to inoculum (sludge from an anaerobic wastewater treatment plant) ratio of 90:10 by volume. The membrane was operated under constant cross flow of nitrogen gas at 2 L/min. Permeate rate was incrementally increased in steps of 2 LMH, terminating at a final flux of 14.2 LMH. Experiment 2 comprised a substrate to inoculum ratio of 80:20 by volume. The membrane was sparged with 3.5 L/min of nitrogen gas. Permeate rate was incrementally increased in steps of 4 LMH, with a final flux of 16.9 LMH.

TS concentrations were 1950±250 mg/L, and flux step durations were 15 minutes. Particle size was measured in Experiment 2 using a Malvern Mastersizer. Mean particle size (50th percentile) was measured at 60 µm. The same particle size was assumed in Experiment 1 as both experiments used substrate and inoculum from the same source.

Experimental conditions in Experiments 1 and 2 are summarised in Table 4-2.

**Table 4-2: Summary of experimental conditions**

<table>
<thead>
<tr>
<th></th>
<th>Experiment 1</th>
<th>Experiment 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas sparge rate [L/min]</td>
<td>2</td>
<td>3.5</td>
</tr>
<tr>
<td>Flux step size [LMH]</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>Final flux [LMH]</td>
<td>14.2</td>
<td>16.9</td>
</tr>
<tr>
<td>Flux step duration, (\theta_f) [min]</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>TS concentration [mg/L]</td>
<td>1700</td>
<td>2200</td>
</tr>
<tr>
<td>Mean particle diameter [µm]</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>Temperature [°C]</td>
<td>34</td>
<td>34</td>
</tr>
</tbody>
</table>
4.4 RESULTS AND DISCUSSION

4.4.1 Visualisation of membrane fouling in 2D

The shear intensity profiles expressed on the membrane surface at 2 L/min and 3.5 L/min are shown in Figures 4-3a and 4-3b, respectively. Qualitatively, the results show a marked difference in membrane shear intensity between the two gas flow rates. The shear profiles follow a typical pattern of decreasing in intensity with increased proximity from the sparger unit. At a gas sparge rate of 2 L/min, membrane shear intensity ranges from 3.6 s$^{-1}$ to 13.6 s$^{-1}$. This gradient in shear is reduced at the higher sparge rate of 3.5 L/min, with minimum shear intensity increasing to 6.2 s$^{-1}$ but maximum shear intensity remaining unchanged at 13.3 s$^{-1}$. Although maximum shear has not increased at the higher gas flow rate, qualitative assessment of the two profiles show that a larger proportion of the membrane receives higher shear at 3.5 L/min than at 2 L/min. Figures 4-3c and 4-3d display cake mass accumulated at 2 L/min and 3.5 L/min, respectively and follow expected behaviour with areas of higher shear experiencing less fouling. At 2 L/min, cake mass ranges from a minimum of 3.7 g/m$^2$ at the bottom of the membrane to 6.3 g/m$^2$ at the top. The higher overall shear produced at 3.5 L/min results in a lower and more even cake formation (2.2 g/m$^2$ to 3.2 g/m$^2$).
4.4.2 Model calibration and validation

The model was calibrated against data in Experiment 1 (Figure 4-4a) and validated with data in Experiment 2 (Figure 4-4b). During the model calibration exercise, the compressibility parameter, $P'$, was estimated at $870 \pm 80$ Pa. While this value is in the low range of $P'$ values reported in literature (1 kPa to 20 kPa) [13, 14], variation is likely due to the difference in sludge types and sources, and the importance of this parameter as identified here, and in the references cited above, indicate it is important to assess system sensitivity to compressibility, particularly where operation is predicted to be close to critical flux.

Figures 4-4a-d display measured vs. simulated results for sparge rates of 2 L/min (Experiment 1) and 3.5 L/min (Experiment 2), respectively, and show good model fit under both conditions. These results indicate that the model effectively simulates the dynamic interaction between flux and pressure, and can also sufficiently replicate flux-step experiments.
Figure 4-4: Experiment vs. simulation: TMP vs. time at 2 L/min (a) and 3.5 L/min (b); dP/dt vs. flux at 2 L/min (c) and 3.5 L/min (d).

4.4.3 Effect of non-uniform planar shear

Due to the capability of this model to simulate 2D shear, cake and flux profiles; it is particularly suited for cases where the membrane experiences variable shear not just along its length but also its width. It is common to encounter non-uniform membrane shear in cases where the sparger unit does not provide full coverage of the membrane (e.g. gas sparged through a single orifice, distance between sparger unit and membrane is too great) or when a plate sparger becomes blocked with biomass and/or precipitated salt. This capability was tested in two cases where the membrane would experience non-uniform planar shear; i) with a single orifice sparger located centrally below the membrane and; ii) with a plate sparger similar to experiment 2, but with only 23% of the sparger active at the right hand side of the unit, i.e. 200 mm blocked, 60 mm unblocked. Both sparger units were located 20 mm from the bottom of the membrane. The overall gas flow rate for both scenarios was set at 3.5 L/min, similar to Experiment 2.
Simulated shear and corresponding cake profiles at these scenarios are displayed in Figures 4-5a to 4-5d. The membrane experiences shear where it overlaps gas flow; i.e., at membrane areas immediately above the single orifice sparger (Figure 4-5a) and above the unblocked portion of the plate sparger in (Figure 4-5c). This leads to cake profiles as shown in Figs. 4-5b and 4-5d, with higher shear regions comprising lower accumulated cake mass (~2 g/m$^2$ in both cases) and the negligible shear region comprising the majority of accumulated cake (~9 g/m$^2$ in both cases). In contrast, the shear profile at full sparger coverage in Experiment 2 (Figure 4-3b) is more uniform along membrane width (x-axis) with all membrane sections experiencing at least 5 s$^{-1}$ shear intensity, and therefore comprising a more uniform cake thickness (Figure 4-3d).

Figure 4-5e displays the change in TMP with step-wise increase in flux over time at the above scenarios and at uniform sparging (fully functional sparger in Experiment 2). At low subcritical flux, the impact of non-uniform sparging on TMP is minimal, but at supercritical flux, TMP increases at a faster rate than with uniform sparging. Figure 4-5f displays the rates of TMP increase (fouling rates), at the non-uniform and uniform sparging scenarios and demonstrates that final non-uniform fouling rates reach 2 (single orifice) to 4 (blocked sparger) times the fouling rate at uniform sparging (unblocked sparger). This can be attributed to a lower active membrane area, defined here as the percentage of membrane receiving > 5 s$^{-1}$ shear intensity, in the single orifice (39%) and blocked plate sparger (30%), than in the uniform gas coverage scenario (100%). Critical flux, or the flux where a deviation from linearity in dP/dt is observed, decreases from approximately 12 LMH in uniform shear to 8.5 LMH in the blocked plate sparger.

The membrane experiences the highest area-average shear intensity with the single orifice sparger (8.4 s$^{-1}$), followed by the unblocked (7.4 s$^{-1}$) and blocked gas spargers (6.0 s$^{-1}$). It is interesting to note that membrane performance is lower with the single orifice sparger than with the unblocked sparger, despite receiving higher area-average shear. This therefore suggests that increasing shear intensity alone may be insufficient for fouling control if membrane coverage by the sparger unit is poor.

The extent of membrane coverage by the sparger is important as it directly relates to the extent of active membrane surface. Model simulations at various non-uniform shear profiles, with overall gas flow rate maintained at 3.5 L/min, revealed a non-linear (power-law) relationship between active membrane area and fouling rate. This profile is displayed in Figure 4-6 at 12 LMH only, for clarity. Confidence intervals for the non-linear regression model are displayed as error bars at each point. Fouling rate is unaffected above 80% membrane coverage. This suggests that non-uniform shearing
is acceptable as long as the proportion of inactive membrane is maintained below a certain threshold.

Figure 4-5: Filtration behaviour at non-uniform shear scenarios: a) Shear intensity and b) cake accumulation with a single orifice sparger; c) shear intensity and d) cake accumulation with a heavily blocked plate sparger; e) TMP vs time at uniform and non-uniform shearing; and f) fouling rate at uniform and non-uniform shearing.
4.5 FURTHER MODEL APPLICATIONS AND LIMITATIONS

The model can be additionally used in the study of the effect of key design and operational parameters such as local geometry around the membrane, sparged gas flow rate, operating flux and the introduction of relaxation periods (gas sparging at no flux); on membrane performance and for the identification of their optimum values for the maximisation of membrane life. This model will also be highly useful in a cost benefit analysis as it allows for dynamic and realistic operational analysis, and can be evaluated in long-term simulations to identify longer term strategies (under a dynamic regime).

A key advantage of the model approach outlined here is the production of a two-dimensional membrane shear profile, which can be scaled up or down proportional to the gas sparging intensity; therefore avoiding repeated CFD modelling and hence saving time and computational resources. The CFD model however, has to be resimulated for different MBR configurations as membrane shear is highly dependent on local geometry. This model also requires further mechanical considerations when modelling hollow fibre membranes; namely the effect of fibre movement in dislodging accumulated cake in addition to membrane shear [10]. Model approach for hollow fibre membranes is discussed further in Chapter 5.

The main limitations are the additional software and technical requirements in simulating the shear profile through CFD, but decoupling this from the membrane flux simulations allows for simulation of dynamic conditions without necessarily dynamic coupling with the CFD model. For example, it
is perfectly appropriate to use CFD to simulate a sparged shear profile, and non-sparged shear profile, and then switch between these two profiles to simulate the impact on membrane fouling dynamically in conjunction with other strategies such as relaxation, which can also be simulated using this model.

4.6 CONCLUSIONS
The extensions to previous models presented here allow for the visualisation of two dimensional shear and cake profiles on a flat sheet membrane surface, therefore allowing analysis of scenarios with non-uniform shear on the membrane surface. In addition, the incorporation of a flux-pressure feedback loop in the model allowed for the simulation of flux-step experiments and the prediction of critical flux. This means inherent model parameters (such as cake compressibility and specific resistance) or complex shear phenomenon, such as in hollow fibre membranes can be estimated from dynamic data. This model can also be expanded for use in true operational scenarios to model long term pressure profiles, and can therefore be particularly useful in design and optimisation studies.
4.7 REFERENCES

5. FURTHER MODEL BASED ANALYSES

ABSTRACT

The membrane fouling model described in Chapter 4 is used as a tool for further analyses; in particular for estimation of shear in complex hollow fibre (HF) membranes and to study the effect of operating parameters (total solids and sparge rate) and filtration strategy, on membrane fouling performance. Shear enhancement due to fibre movement and fibre contact in hollow fibre membranes is difficult to measure and predict in multiple fibre configurations due to the complex fibre geometry and numerous controlling factors, whose combined effect on shear is difficult to quantify and correlate. In this chapter, fibre-induced shear enhancement is represented by a single parameter, \( \omega \), and estimated for the pilot-scale HF anaerobic membrane bioreactor (AnMBR) by model calibration against short-term flux-step experiments. In total solids (TS) concentration of 2 g/L, \( \omega \) of 5 times CFD shear was identified. Shear enhancement was not identified in 35 gTS/L (\( \omega = 1 \)) suggesting fibre movement is constrained in concentrated sludge. The model identified an optimum gas sparge rate of 10 L/min in 2 gTS/L to 20 gTS/L. Long filtration periods at lower flux is a more beneficial strategy over a short filtration period and higher flux. Model predictions identified a linear relationship between required membrane surface area and sparger gas volume per day, and tank volume with approximately 5 HF membrane modules and 76 m\(^3\)/day of gas required per m\(^3\) tank volume. Therefore, the membrane fouling model developed in this thesis has the capability of quantifying complex membrane mechanisms, such as fibre shear, and can be used for the detailed design and optimisation of AnMBRs.
5.1 INTRODUCTION

The previously developed membrane fouling model in Chapter 4 incorporates various interacting factors affecting membrane fouling into an integrated tool that allows for determination of platform independent fouling characteristics and a better fundamental understanding of controlling mechanisms. The model also allows for quantification of complex fouling parameters that cannot easily be measured in-situ, as was demonstrated in Chapters 3 and 4 when estimating cake compressibility of model foulants and slaughterhouse wastewater. In this chapter, the model is used as a tool for further analyses; in particular to estimate shear in complex membrane geometry and to study the effect of operating parameters (total solids and sparge rate) and filtration strategy, on membrane fouling performance.

In configurations where the membrane can flex, such as the hollow fibre (HF) membrane, fouling control mechanisms are more complex than in rigid flat sheet (FS) membranes and can be enhanced by fibre-fibre interactions. In addition to bubble-induced shear, fouling in gas-sparged HF membranes is controlled by fibre oscillation and fibre-fibre contact [1]. Lower fouling rates in oscillating fibres[2, 3] are attributed to an enhancement of shear due to fibre oscillation; which can be comparable in magnitude to shear induced by gas sparging alone [4]. In HF modules with multiple fibres held together in a high packing density, oscillating fibres can collide, creating shear forces that can be an order of magnitude greater than in single gas-sparged fibres [5-7].

Estimating the hydrodynamics in a multiple fibre bundle is however; a complex modelling and experimental challenge due to the presence of the numerous and randomly oscillating fibres. Potential computational fluid dynamics (CFD) modelling strategies could include; representations of the moving multifibre HF as a moving mesh; or by a two-way fluid-structure interaction which requires coupling of an external structural physics solver with the CFD solver. These approaches however are highly case specific and complex to configure, and require considerable computational resources and time, especially in the simulation of the movement of hundreds of fibres. Experimental estimation of fibre shear has used electrochemical shear probes [5, 6] and direct particle image velocimetry [8]. Shear profiles obtained from these studies are however, only representative for the set-ups and conditions used in the experiments. Studies using these techniques also identified problems during shear estimation in bubbly-flow and at high fibre oscillation due to the highly variable and complex nature of bubble and fibre movement.

Experimental studies have identified various key parameters of fibre-induced shear, including fibre characteristics such as fibre tightness, length, diameter and packing density; and operating conditions such as reactor total solids (TS) concentration (which affects liquid viscosity) and gas
sparge rate [2, 5-7, 9-11]. In general, enhanced shear and hence lower fouling rates have been observed in conditions favouring a greater degree of fibre oscillation (fibre amplitude and oscillation frequency); such as in fibres with <100% tightness (distance between fixed ends/fibre length × 100) [2, 8, 9, 12]; in fibres that were longer and thinner [2, 10]; at low operating TS (viscosity) [2] and at high gas sparge rates [2, 5]. High packing density encourages fibre-fibre contact and increases membrane shear; however there will exist an optimum packing density, dependant on membrane configuration and wastewater characteristics, above which internal fibre clogging will be amplified [11]. The various and interrelated factors affecting multifibre shear and fouling are generally studied in isolation, and a combined effect of these factors is difficult to quantify and correlate based on the above studies.

Fibre-induced shear enhancement is treated here as a single parameter and estimated for the pilot-scale HF AnMBR using the membrane fouling model in Chapter 4. The HF bundle is represented in the CFD model as a rigid cylindrical wall over which surface shear is imparted. Fibre-induced enhancement of CFD shear is estimated by calibrating the model against short-term flux-step experiments conducted on the pilot-scale HF AnMBR using raw and digested slaughterhouse wastewater. Fibre-induced shear enhancement, represented by a multiplier, \( \omega \), is estimated via statistical non-linear least squares parameter estimation.

Model based analyses of the combined effect of TS and sparge rates are useful in design, optimisation and cost-benefit analyses; and provide information that can be used in scale-up of the reactor. A key advantage in both aerobic and anaerobic submerged membrane bioreactors (SMBRs) is the ability to maintain high TS concentrations and hence biomass inventory; therefore increasing treatment capacity at a lower reactor footprint [13]. However, the increased sludge viscosity at high TS will increase energy required for sufficient shearing of the membrane. High TS also increases the probability of foulant deposition on the membrane [14]. Hence, the benefits of high treatment capacity in an AnMBR need to be weighed against the higher fouling propensity at high TS.

Operating gas sparge rate directly affects membrane fouling via imparted membrane shear. Membrane shear in dual-phase bubbly flows can be higher than in single-phase crossflow [5] due to the combined effects of bubble induced liquid crossflow and high localised velocity gradients and eddies in bubble-wakes. Sparging however also has associated energy costs. In AnMBRs, biologically produced biogas is recycled for gas sparging hence reducing (and maybe eliminating) the requirement for external gas sources. However, the use of energy-rich biogas for gas sparging of the membrane affects the energy recovery potential of the AnMBR. The energy requirement associated with gas recirculation also increases at higher sparge rates. Hence, model based fouling
predictions will enable the identification of an optimum gas sparge rate for fouling control in a given system.

Long term membrane performance can also be affected by operating filtration strategy such as duration of filtration and inclusion of relaxation and backwash. The model is used here to identify the effect of operating at a lower flux for a longer period (shorter relaxation time) versus higher flux for a shorter period (longer relaxation time) at similar overall process hydraulic retention times (HRTs).

Membrane performance is determined and compared for each scenario via the response of transmembrane pressure (TMP) with stepping-up of applied membrane flux. This enables calculation of fouling rates (dP/dt) and critical flux, defined here as the flux at which dP/dt >0.01 KPa/min [15].

5.2 MODEL OVERVIEW

A brief overview of the model is presented in Figure 5-1; showing the relationship between key factors affecting membrane fouling behaviour. Parameters explored in this chapter, including TS concentration, gas sparge rate and fibre-induced shear enhancement are highlighted in red.

Figure 5-1: Model overview. Parameters explored in this chapter are highlighted in red.
**CFD model prediction of membrane shear**

A two fluid (Eulerian-Eulerian) approach was used to simultaneously model liquid and gas phases. The sludge mixture was treated as a homogenous single phase and total solids (TS) concentration was linked with rheology. CFD shear was estimated for pilot-scale scenarios containing TS concentrations of 2 gTS/L, 10 gTS/L, 20 gTS/L and 35 gTS/L.

At 2 gTS/L, Equation 2-55 was used to estimate viscosity. At 10 gTS/L and 20 gTS/L, Casson viscosity correlations (Equations 2-56 to 2-58). At the higher TS concentration of 35 g/L, a non-Newtonian Bingham model was applied (equation 5-1). Values of Bingham model parameters, \( \tau_B \) and \( \mu_B \), were taken from Battistoni et al’s (1997)[16] measurements on mesophilic (33°C) anaerobically digested sludge with a TS of 35 g/L.

\[
\tau = \tau_B + \mu_B \gamma 
\]  

(5-1)

Where:

- \( \tau \) = Shear stress [Pa]
- \( \tau_B \) = Bingham yield stress = 0.4 Pa
- \( \mu_B \) = Bingham viscosity consistency = 0.008 Pa.s

Bubble size is linked with gas flow rate according to Bowers’ empirically obtained correlations [17]. At low gas flow rate (<10 L/min) bubble size was determined using Equation 2-51. At high gas flow rate (>20 L/min) the bubble diameter was calculated using Equations 2-52 and 2-53, for laminar and turbulent conditions respectively. An average of laminar and turbulent bubble size was incorporated into the CFD model. Bubble break up and coalescence was not considered in this study.

Fluid turbulence was modelled using a k-\( \varepsilon \) turbulence model, which is the most widely validated of the existing Reynolds Stress turbulence models [18, 19]. Default values of the empirical turbulence constants are used in the CFD analysis, in accordance with the standard values provided by Rodi [18]. Additional models include the use of a Grace drag model [20] for gas-liquid momentum transfer, Tomiyama lift force model [21] and Sato’s model [22] for turbulence enhancement in bubbly flows. Further details of CFD submodels are detailed in Chapter 2.
The AnMBR domain was meshed using ANSYS meshing software. The mesh comprises a total approximately 342,000 elements and is within quality recommendations (orthogonal quality >0.1, skewness <0.95). The solver was run in steady-state mode for at least 100 iterations or until constant values of monitored parameters (fluid velocity, maximum and average wall shear) were reached. The maximum residual target was $1 \times 10^{-3}$.

**Fouling model**

A sectional fouling model was used here to predict dynamic cake formation on the membrane as described in Chapter 4. The membrane surface is divided into a number of sections over which cake accumulation rate, $dM_c/dt$, is calculated (Equation 4-2). Shear profiles over the HF membrane typically vary lengthwise. Hence, the cylindrical HF membrane was divided into 8 longitudinal sections around its surface. CFD shear was ‘sampled’ at 100 points on each line and averaged radially to obtain a single longitudinal shear vector.

The increase in resistance due to cake accumulation leads to an increase in transmembrane pressure (TMP) which is dynamically linked to applied flux, enabling the simulation of flux-TMP behaviour and hence the prediction of critical flux. The model was validated with FS membranes against flux-step experiments on 2 g/L digested slaughterhouse wastewater (Chapter 4). In this chapter, HF filtration tests were also conducted on slaughterhouse wastewater and hence model parameter values are the same as in Chapter 4, with the exception of measured HF membrane resistance, $R_m$, and the addition of a model estimated shear enhancement coefficient, $\omega$.

A weak form of the critical flux definition is applied here, which defines $R_m$ as the resistance below critical flux (i.e. $dP/dt < 0.01$ KPa/min). The shear enhancement coefficient, $\omega$, comprises additional fibre-induced shear which is not included in the CFD model. The coefficient, $\omega$, was estimated via parameter estimation during calibration of the model against experiments using a non-linear least squares parameter estimation method (Levenberg-Marquardt algorithm) with TMP as fitted output and residual sum of squares ($J$) as objective function. Parameter uncertainty was estimated from a two-tailed t-test (95% confidence interval) on the parameter standard error, which was calculated from the objective parameter Jacobian at the optimum (i.e linear estimate) [23]. Pore fouling was neglected, as previous model validation and experiments (in Chapter 3 and 4) confirmed a cake-fouling tendency of high protein-containing slaughterhouse wastewater.

Table 5-1 summarises model parameters specific to this chapter. A full list of model parameters is detailed in Table 4-1.
Model based $\omega$ estimation was conducted during model calibration against results from 6 batch filtration experiments on a pilot scale HF AnMBR treating slaughterhouse wastewater; which was comprised of a 200 L tank (0.47 m diameter by 0.778 m wetted height) containing a vertically mounted submerged HF module (Zenon ZW-10, approx. 600 mm height and 100 mm diameter, 0.93 m$^2$ surface area). The HF module comprises approximately 300 fibres arranged in a bundle around a central gas line column in a packing density of 10 fibres per cm$^2$. Each fibre is approximately 0.55 m long and has a diameter of 1.9 mm. The distance between the fixed ends of the fibres was 0.535 m. Hence fibre tightness in this module was approximately 97.3%. The membrane is sparged with nitrogen gas via a coarse bubbler (four 2 mm diameter holes) located at the bottom of the fibre bundle.

### Table 5-1: Fouling model parameters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Value</th>
<th>Units</th>
<th>Notes/References</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_m$</td>
<td>Membrane resistance</td>
<td>1-2.5 ($\times 10^{12}$)</td>
<td>m$^{-1}$</td>
<td>Measured</td>
</tr>
<tr>
<td>$d_p$</td>
<td>Particle diameter</td>
<td>30-80</td>
<td>µm</td>
<td>Measured</td>
</tr>
<tr>
<td>$n_{i,j}$</td>
<td>Number of membrane sections</td>
<td>100</td>
<td></td>
<td>Set</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>32-35</td>
<td>°C</td>
<td></td>
</tr>
<tr>
<td>$\omega$</td>
<td>Shear enhancement factor</td>
<td></td>
<td></td>
<td>Estimated</td>
</tr>
</tbody>
</table>

### 5.3 EXPERIMENTS

Model based $\omega$ estimation was conducted during model calibration against results from 6 batch filtration experiments on a pilot scale HF AnMBR treating slaughterhouse wastewater; which was comprised of a 200 L tank (0.47 m diameter by 0.778 m wetted height) containing a vertically mounted submerged HF module (Zenon ZW-10, approx. 600 mm height and 100 mm diameter, 0.93 m$^2$ surface area). The HF module comprises approximately 300 fibres arranged in a bundle around a central gas line column in a packing density of 10 fibres per cm$^2$. Each fibre is approximately 0.55 m long and has a diameter of 1.9 mm. The distance between the fixed ends of the fibres was 0.535 m. Hence fibre tightness in this module was approximately 97.3%. The membrane is sparged with nitrogen gas via a coarse bubbler (four 2 mm diameter holes) located at the bottom of the fibre bundle.
Figure 5-2: Experimental setup of the pilot scale HF AnMBR

Filtration or flux-step experiments were conducted in accordance with the protocol described by Le Clech et al (2003) [24]. Flux was maintained and controlled using a peristaltic pump on the permeate stream. Pressure change was continuously monitored by means of a pressure transducer (model Druck PTX 1400, GE) situated at the permeate stream. The permeate stream was recirculated into the bioreactor to maintain the liquid level within the reactor. Furthermore, temperature was measured via an RTD sensor (model SEM203 P, W&B Instrument Pty.), and was controlled at the operating temperatures of the AnMBR (32-37 °C). Both pressure transducer and temperature output signal (4-20 mA) were logged via PLC onto a computer.

The experiment HF-3.5WW was conducted using the same wastewater and sparger conditions as the FS Experiment 2 in Chapter 4; i.e. in digested slaughterhouse wastewater (batch digested for 65 days) with total solids concentration and nitrogen gas sparge rate maintained at 2.2 g/L and 3.5 L/min, respectively. This enabled a direct comparison of FS and HF performance. Permeate flux was increased in step sizes of 4 LMH until a maximum flux of 25 LMH. Results from the flat sheet Experiment 2 in Chapter 4 are presented here for comparison purposes and is renamed FS-3.5WW for easier discussion.

Experiments HF-2RS1 and HF-3.5RS1 were conducted on combined slaughterhouse wastewater (i.e. substrate only without inoculum), with sparge rates of 2 L/min and 3.5 L/min, respectively. Wastewater in both experiments were sourced from the combined wastewater of an Australian
cattle processing facility at the same time (3\textsuperscript{rd} July 2012) and hence comprised similar wastewater characteristics (2.4 gTS/L, chemical oxygen demand (COD) of 3.1 g/L and total protein of 1.1 g/L). Flux was increased in steps of 4 LMH until a maximum flux of 17 LMH in HF-2RS1 and 25 LMH in HF-3.5RS1.

Experiments HF-3.5RS2 (32°C) and HF-3.5RS2 (24°C) were conducted at 32°C and 24°C, respectively on combined slaughterhouse wastewater sourced at the same time (17\textsuperscript{th} July 2012) with 2.8 gTS/L, 3.8 gCOD/L and total protein of 2.1 g/L. Nitrogen sparge rates in both experiments were maintained at 3.5 L/min. Flux was increased in steps of 4 LMH until a final flux of 20 LMH.

Experiment HF-30S was conducted on AnMBR sludge after 200 days continuous mesophilic operation at an Australian cattle processing facility [25]. The sludge had a TS concentration of 34.6 g/L. Permeate flux was increased in step sizes of 2 LMH until a maximum flux of 11 LMH. Nitrogen sparge rate during the flux-step experiment was maintained at a high flow rate of 30 L/min; to maximise membrane shear in the high solids environment.

Particle size was measured using a Malvern Mastersizer. Mean particle size (50\textsuperscript{th} percentile) was 30 µm in raw slaughterhouse wastewater and 60 µm in digested slaughterhouse wastewater. Particle size was not measured for AnMBR sludge in experiment HF-30S, and was assumed to equal 80 µm based on measurements on sludge from a later mesophilic operation of the AnMBR.

Membranes were systematically cleaned prior to each measurement. Cleaning protocol included physical cleaning in tap water (high air flowrate sparging in water) and a soft sponge, after which they were soaked in 0.5 wt% sodium hypochlorite and subsequently 0.2 wt% citric acid for 2 hours each. The membranes were rinsed again and stored in tap water until the next experiment. Clean water permeability was measured subsequently to evaluate permeability recovery prior to the next trial.

Experimental conditions are summarised in Table 5-2.
### Table 5-2: Summary of filtration experiments

<table>
<thead>
<tr>
<th></th>
<th>Dilute wastewater</th>
<th>Concentrated sludge</th>
</tr>
</thead>
<tbody>
<tr>
<td>WW source</td>
<td>FS-3.5WW</td>
<td>HF-3.5WW</td>
</tr>
<tr>
<td></td>
<td>HF-2RS1</td>
<td>HF-3.5RS1</td>
</tr>
<tr>
<td></td>
<td>HF-3.5RS2 (32°C)</td>
<td>HF-3.5RS2 (24°C)</td>
</tr>
<tr>
<td></td>
<td>HF-30S</td>
<td>HF-3.5RS2</td>
</tr>
<tr>
<td>Gas sparge rate [L/min]</td>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>Flux step size [LMH]</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>Final flux [LMH]</td>
<td>17</td>
<td>25</td>
</tr>
<tr>
<td>Flux step duration [min]</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>TS [g/L]</td>
<td>2.2</td>
<td>2.2</td>
</tr>
<tr>
<td>Temperature</td>
<td>32</td>
<td>32</td>
</tr>
<tr>
<td>Mean floc particle diameter [µm]</td>
<td>60</td>
<td>30</td>
</tr>
</tbody>
</table>

1. after 65 days batch digestion mesophilic (32-35°C)
2. Total chemical oxygen demand (COD) = 3.1 g/L; total protein = 1.1 g/L;
3. Total COD = 3.8 g/L; total protein = 2.1 g/L
4. after 200 days continuous mesophilic operation
5.4 RESULTS AND DISCUSSION

5.4.1 Fibre-induced shear enhancement

*Identification of shear enhancement*

FS-3.5WW and HF-3.5WW are directly comparable as they were conducted in similar conditions; i.e. the same digested slaughterhouse wastewater and sparge rate. Therefore, differences in membrane fouling performance can be directly linked to the differences in configuration between the two membranes.

Figure 5-3 illustrates CFD shear profiles in dilute wastewater (~2 gTS/L) in the HF configuration (planar FS shear is shown in Figure 4-3b). The HF membrane is modelled as a rigid cylindrical wall and hence CFD shear only comprises bubble-induced surface shear.

![Shear Strain Rate](image)

Figure 5-3: CFD shear profile on the HF membrane at 2 gTS/L and gas sparge rate of 3.5 L/min.

In the HF AnMBR at a sparge rate of 3.5 L/min, shear rates range from 4.9 to 10.1 s⁻¹. At a sparge rate of 3.5 L/min, the FS AnMBR receives a higher shear rate (6.2 -13.3 s⁻¹) than the HF AnMBR. Table 5-3 summarises CFD shear rates at the scenarios studied.
Table 5-3: Summary of CFD shear in the flat sheet

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Total solids [g/L]</th>
<th>Sparge rate [L/min]</th>
<th>Membrane averaged shear rate [s⁻¹]</th>
<th>Min. shear rate [s⁻¹]</th>
<th>Max shear rate [s⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat sheet</td>
<td>2</td>
<td>3.5</td>
<td>7.4</td>
<td>6.2</td>
<td>13.3</td>
</tr>
<tr>
<td>Hollow fibre</td>
<td>2</td>
<td>3.5</td>
<td>6.5</td>
<td>4.9</td>
<td>10.1</td>
</tr>
</tbody>
</table>

TMP profiles and corresponding fouling rates for FS-3.5WW and HF-3.5WW are shown in Figures 5-4a and 5-4b, respectively. Membrane flux is displayed as a normalised flux per square metre membrane area, L/m²-h or LMH, to allow direct comparison between the different membranes and scenarios. The HF membrane had a higher critical flux (10 LMH) than the FS membrane (7 LMH) despite the CFD model predicting a higher average shear for the FS membrane (Table 5-3), confirming the presence of an additional mechanism of fouling control in the HF which is most likely fibre-induced enhanced shear that is missing in the CFD model.
Figure 5-4: TMP vs. time and corresponding dP/dt for a) FS-3.5WW and b) HF-3.5WW

Estimation of shear enhancement in the HF membrane

The shear enhancement coefficient, $\omega$, was estimated during model calibration against the dilute wastewater filtration experiments (refer to Table 5-2). Measured and simulated (after calibration) TMP vs. time and corresponding fouling rates (dP/dt) are shown in Figure 5-4b for HF-3.5WW and in Figure 5-5 for the remaining dilute wastewater experiments.
Figure 5-5: Measured vs. simulated TMP vs. time and corresponding dP/dt for a) HF-2RS1; b) HF-3.5RS1; c) HF-3.5RS2 at 32°C and d) HF-3.5RS2 at 24 °C
Estimated values of \( \omega \) for each experiment are summarised in Table 5-4; with a consistent estimate of 5 times CFD predicted shear. This indicates that shear enhancement is not substantially influenced by concentration or temperature (Note: HF has 97% tightness)

**Table 5-4: Summary of model estimated fibre shear factor, \( \omega \)**

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fibre shear factor, ( \omega )</th>
</tr>
</thead>
<tbody>
<tr>
<td>HF-3.5WW</td>
<td>5.06 ± 0.0008</td>
</tr>
<tr>
<td>HF-2RS1</td>
<td>5.00 ± 0.0015</td>
</tr>
<tr>
<td>HF-3.5RS1</td>
<td>5.50 ± 0.0004</td>
</tr>
<tr>
<td>HF-3.5RS2 (32°C)</td>
<td>5.44 ± 0.115</td>
</tr>
<tr>
<td>HF-3.5RS2 (24°C)</td>
<td>5.00 ± 0.098</td>
</tr>
</tbody>
</table>

Figure 5-6 compares predicted fouling rates with and without shear enhancement; i.e. at \( \omega \) of 5 and 1, respectively, with a reactor TS of 2 g/L and gas sparge rate of 3.5 L/min. At \( \omega \) of 5 and 1 respectively, critical fluxes are 9 LMH and 5 LMH. Hence assuming 100% fibre tightness at \( \omega = 1 \) and 97% fibre tightness at \( \omega = 5 \), a 3% looser fibre creates an improvement in critical flux of 1.8 times.

![Figure 5-6: Fouling rates with (\( \omega = 5 \)) and without (\( \omega = 1 \)) shear enhancement](image)

Figure 5-6: Fouling rates with \((\omega = 5)\) and without \((\omega = 1)\) shear enhancement
Effect of high TS concentration on enhanced fibre-shear

With concentrated sludge at 35 gTS/L the model showed good fit with experimental values (Figure 5-7) without enhancement of CFD shear (i.e. $\omega = 1$), even at a high sparge rate of 30 L/min; hence suggesting a lack of fibre contribution to shear in concentrated sludge.

Figure 5-7: a) TMP vs. time in concentrated sludge (35 gTS/L) and b) corresponding fouling rate

Wicaksana et al (2006) [2] observed a decline in fibre amplitude and frequency when viscosity was increased from 0.001 Pa.s (i.e. water) to 0.003 Pa.s (equivalent to 15 gTS/L), therefore suggesting that the high viscosity at high TS concentrations impedes fibre movement. Sludge at 35 gTS/L, will have a non-Newtonian rheology, with viscosity dependant on applied shear. At a maximum membrane shear of 92 s$^{-1}$ experienced by the membrane at 30 L/min, the viscosity near the membrane is approximately 0.012 Pa.s; or 12 times water viscosity; hence it seems reasonable that the amplitude and frequency of fibre movement would be reduced under these conditions to the point where there is no observable benefit in terms of improved shear.

Visual appraisal of membrane fouling (after 200 days continuous mesophilic operation) shows that the major fouling zones (Figure 5-8b) on the HF membrane correspond well with CFD predicted shear (Figure 5-8a); i.e. membrane zones with high shear correspond with zones of low cake accumulation and vice versa.
The relationship between critical flux and sparge rate at TS of 2, 10 and 20 g/L is shown in Figure 5-9, for the pilot-scale HF AnMBR. Critical flux at all TS concentrations plateau after a gas sparge rate of 10 L/min, with maximum critical flux achieved at around 20 L/min gas sparge rate. The existing reactor and membrane configuration has maximum critical fluxes of 8.5 LMH, 5 LMH and 4 LMH at respective TS concentrations of 2 g/L, 10 g/L and 20 g/L. These relationships were obtained without considering fibre-induced shear enhancement, as $\omega$ could not be estimated due to lack of flux-step data in the intermediate TS of 10 g/L and 20 g/L. Hence actual critical fluxes may be higher than those shown in Figure 5-9 due to fibre-induced shear, with degree of shear enhancement decreasing as TS increases. Regardless, the general trends in critical flux behaviour with increasing sparge rate will remain the same.
Application to the real system

Biological treatment capacity of pilot-scale AnMBR sludge has an upper organic loading rate (OLR) limit of approximately 0.14 gCOD/gTS-day (3.5gCOD/L-day with a sludge inventory of 25 gTS/L) [25]. At an average feed chemical oxygen demand (COD) of approximately 6 g/L [25]; and a sludge inventory of 20gTS/L, an HRT of 2 days is required for sufficient biological treatment. In the pilot-scale AnMBR (150 L reactor volume and 0.93m$^2$ HF membrane surface area) a 2 d HRT requires an operating membrane flux of 3 LMH and can be maintained at an operating gas sparge rate of approximately 6 L/min, according to Figure 5-9, assuming a tight fibre membrane with little to no fibre movement and hence no shear enhancement.

To achieve an HRT of 2 days at larger tank volumes, larger membrane surface areas will be required to maintain operating flux below the membrane’s critical flux. Figure 5-10a shows the number of HF modules (with similar local geometry to the pilot-scale ZW-10 and each with a surface area of 0.93m$^2$) required as tank volumes are increased. Corresponding gas volumes required (per day) for membrane sparging is shown in Figure 5-10b. The relationship between tank volumes and required membrane area and gas volume is linear. Least squares regression with 95% confidence level finds that approximately 5 HF membrane modules and 76 m$^3$/day of gas will be required per m$^3$ tank volume. This information can directly feed into cost-analyses during scale-up design and for the estimation of capital expenditure (CAPEX), including membrane costs, and operating expenditure (OPEX), including costs associated with sparging.
Figure 5-10: a) No. of HF modules required per m$^3$ reactor volume; b) Sparger gas volume (m$^3$) required per m$^3$ reactor volume.

5.4.3 Effect of filtration strategy

Model estimated cake mass distribution is shown in Figure 5-11 at two of the filtration strategies used on-site during continuous operation at 35 gTS/L and 30 L/min gas sparge rate. These include filtering at 6 LMH over 12 hours and 3 LMH over 24 hours; each having a respective average cake mass of 2.8 g/m$^2$ and 2.0 g/m$^2$. Relaxation effects on the membrane were not included in the model. Therefore, without sacrificing operating hydraulic retention times (HRT), lower membrane fouling and hence improved membrane performance can be achieved if the membrane is operated at half the filtration flux over twice the filtration period.

Figure 5-11: Model estimated cake mass at two filtration strategies - 6 LMH over 12 hours and 3 LMH over 24 hours.
5.4.4 Model limitations

While the representation of shear enhancement by a single coefficient calibrated against experiments may limit the model to the conditions used in the experiment, its quantification requires simple short-term flux-step tests, which is still a relatively simple exercise for characterising a complex mechanism such as fibre-induced shear. Future work to increase model applicability can include comprehensive filtration tests at a range of intermediate TS concentrations, fibre characteristics (packing density, length, diameter, tightness etc.) and sparge rates. Model based calibrations against these studies will quantify fibre-induced shear enhancement at each of these varying conditions; and can be correlated into an overall model that can be used to understand the combined effect of the various controlling factors in fibre-induced shear.

HF membranes at high fibre packing density have a tendency towards internal clogging, which the model does not consider. In wastewaters comprising fibrous material, such as grassy, paunch solids in slaughterhouse wastewater, fibre clogging will be an issue that requires consideration. In larger scale membranes with higher packing densities, clogging may also be an issue. Interfibre clogging as a mechanism is complex, and when it does occur, cannot be controlled in-situ even via backwash or chemical cleaning [12]. It will hence be more efficient to design for conditions that mitigate clogging; i.e. use looser, less densely packed fibres and pre-treat wastewater to reduce potential clogging agents [11]; than to control clogging during operation itself. This will reduce the need for considering clogging as mechanism in model based analysis.

5.5 CONCLUSIONS

The HF membrane used in this study had a fibre tightness of 97% and showed greater membrane performance (1.4 times higher critical flux) than the flat sheet (FS) membrane in similar operating conditions, confirming that an additional shearing mechanism exists in the HF membrane which can be attributed to fibre-induced shear. In dilute wastewater with total solids (TS) concentration of 2 g/L, shear enhancement factor ($\omega$) of 5 was identified. Shear enhancement was not identified in concentrated sludge with 35 gTS/L; i.e. $\omega = 1$; suggesting that fibre is constrained in concentrated sludge.

At TS concentrations ranging from 2 to 20 g/L; an optimum gas sparge rate of 10 L/min was identified. In the pilot-scale AnMBR (150 L reactor volume and 0.93m² membrane surface area) maintaining a 2 d HRT (in 20 gTS/L and organic loading rate of 0.14 gCOD/gTS-d) requires an operating membrane flux of 3 LMH which can be sustained at an operating gas sparge rate of
approximately 6 L/min. To achieve an HRT of 2 days at larger tank volumes, larger membrane surface areas will be required to maintain operating flux below the membrane’s critical flux. Model predictions identified a linear relationship between tank volume and required membrane area and sparger gas volume per day, with approximately 5 HF membrane modules and 76 m³/day of gas required per m³ tank volume. The model also predicted lower membrane fouling and hence improved membrane performance at 3 LMH over 24h filtration period compared with 6 LMH over a 12h filtration period. Therefore, without sacrificing operating hydraulic retention times (HRT), lower membrane fouling and hence improved membrane performance can be achieved if the membrane is operated at half the filtration flux over twice the filtration period.

Future work to increase model applicability can include comprehensive filtration tests at a range of intermediate TS concentrations, fibre characteristics (packing density, length, diameter, tightness etc.) and sparge rates. Model based calibrations against these studies will quantify fibre-induced shear enhancement at each of these varying conditions; and can be correlated into an overall model that can be used to understand the combined effect of the various controlling factors in fibre-induced shear.
5.6 REFERENCES

6. CONCLUSIONS AND RECOMMENDATIONS

This work investigated fouling in an anaerobic membrane bioreactor (AnMBR), in particular for the treatment of food and animal processing wastewaters. Particulates and substrate composition had complex effects on membrane fouling, suggesting that slowly degrading substrate in industrial AnMBRs will impact bulk filtration properties. The incorporation of 3D computational fluid dynamics (CFD) into the overall membrane fouling model allowed a direct estimation of the effect of operating strategy and reactor and membrane geometry into membrane fouling analysis. Major conclusions based on specific research objectives are listed below along with significance of the findings as well as recommendations for future work.

6.1 OBJECTIVE 1: HYDRODYNAMIC CHARACTERISATION (CHAPTER 2)

Conclusions:
The chosen CFD approach was successfully validated against literature obtained shear measurements. Minimal differences in velocity and shear profiles between two and three phase approximations of the AnMBR suggested that sludge settling had a negligible effect on hydrodynamics.

Significance and implications of findings:
The incorporation of CFD directly linked membrane shear with operating conditions, such as sparge rate and viscosity (total solids), as well as to internal reactor geometry, therefore enabling the study of these effects on shear during reactor design and optimisation. The multidimensional aspect of 3D CFD modelling enabled the incorporation of a distributed shear parameter into fouling models, which provided a calculation of cake and flux distributions on the membrane surface.

Recommendations for future study:
The CFD model was validated against small-scale lab experiments. Larger and full-scale industrial applications, are expected to have more complex bubble behaviour through the effects of bubble coalescence and transient bubble regimes. Bubble size generally increases with height, due to bubble coalescence and reduction in static pressure and has complex effects on shear. In bubbly flow regimes, larger bubbles will impart lower shear than smaller bubbles. However, bubble coalescence at high gas flowrates can encourage the formation of slug-flow or turbulent churn which are highly unstable and can impart high turbulent shear. Neglecting bubble coalescence can
either overestimate shear in a bubbly regime, or underestimate shear in slug-flow/turbulent churn regimes. These effects will be important in tall reactors containing long membranes, and will affect the upper parts of the membrane where bubble flow is fully developed. Therefore, for application to larger scale AnMBRs, future model approach should include a bubble size distribution, rather than a constant mean bubble diameter, and incorporate the effects of bubble coalescence and break-up.

The CFD model also neglects the role of in-reactor biomass gas production on velocity and shear profiles. In full-scale applications, the effect of biogas production may have a significant effect on membrane shear. Therefore, a natural progression of the model is the incorporation or coupling of biokinetic models such as the anaerobic digestion model no. 1 (ADM1) [1] into the hydrodynamic model. This can be achieved either by incorporating a decoupled biological and solids settling model with the CFD model or using a three-phase CFD approach, with sludge volume fractions directly linked to ADM1 kinetics to describe interphase mass transfer from solids to gas.

The CFD modelling of complex membranes such as hollow fibres will require moving meshes for the numerous fibres; and the CFD solver will require coupling with an external structural physics solver to estimate two-way fluid-structure interactions (FSI). This is currently computationally prohibitive. However, it is not unreasonable to expect major developments in computational capacity and speed in the near future which may make the direct simulation of multifibre shear via two-way FSI feasible.

6.2 OBJECTIVE 2: PARTICULATE FOULING AND THE EFFECT OF SUBSTRATE COMPOSITION (CHAPTER 3)

Conclusions:
Proteins had a significant impact on fouling behaviour, individually, as well as in binary and ternary mixtures with carbohydrates and lipids. Protein aggregation due to thiol interactions and surface charge had an important role in protein fouling. Protein hydrophobicity and compressibility were also key contributors to the high protein fouling propensity. Filter cake compressibility was directly related to protein content; with P’ (pressure at which initial specific cake resistance is doubled) increasing from approximately 830 Pa in 100% protein to 5000 Pa in 50% protein mixtures and 10,000 Pa in 10% protein mixtures. Protein showed high reactivity with lipids causing synergistic fouling behaviour in protein-lipid mixtures and had a surfactant effect on lipids, changing the fouling mechanism from pore fouling (irreversible and irrecoverable) in pure lipids to cake fouling (reversible) in protein-lipid mixtures. AnMBR sludge with P/C/L of 90/10/1 showed protein
dominant fouling behaviour, with similar cake compressibility (870 Pa) and fouling rates (max dP/dt of 0.084 KPa/min) to pure protein (max dP/dt of 0.088 KPa/min).

**Significance and implications of findings:**
Mechanistic model foulant analyses suggest that high particulate protein streams will have higher fouling rates than particulate carbohydrate rich streams. The surfactant effect of protein on lipids suggests that protein addition may be a viable strategy in fatty wastewaters to protect the membrane from long term pore fouling and hence irrecoverable fouling. The trade-off in this strategy will be the higher fouling rates due to protein-lipid synergisms.

Surface modification (to make the surface more hydrophilic) is a widely applied membrane fabrication strategy to mitigate membrane accumulation of hydrophobic organic foulants. However, this study identified that after an initial filtration period, surface characteristics take on that of the fouling layer rather than the membrane, and that surface modification may not be an effective fouling mitigation strategy for particulate-laden waste streams such as food and animal processing wastewater.

**Recommendations for future study:**
Further analysis to identify and characterise protein fouling and protein-lipid interactions is recommended. Traditional analytical techniques such as scanning electron microscopy (SEM) and transmission electron microscopy (TEM) will provide qualitative visualisation of filter cake morphology (structure) and may also detect internal (pore) foulants. A number of advanced analytical techniques exist for the characterisation of protein fouling [2] and a few of these are mentioned here as suggestions for future analysis:

(i) In mixed foulant mixtures, the use of *UV spectrophotometry* will provide an idea of the location of protein foulants on the surface and whether protein dominates in the cake layer.

(ii) Protein denaturation can greatly increase its fouling propensity, and was observed in this thesis during pure protein and protein-carbohydrate filtration. *Attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR)* analysis has been used in literature to identify conformational changes in protein due to denaturation and will be useful in future studies to characterise the effect of denatured proteins on fouling as well as to identify conditions in which denaturation is favoured.
Atomic force microscopy (AFM) can be used to examine foulant morphology in hydrated conditions. AFM can also measure hydrophobic forces and electrical double-layer interactions between proteins and membrane which can be linked to protein fouling propensity.

Model foulants effectively represented the fouling behaviour in high protein and carbohydrate containing AnMBR (digested slaughterhouse) sludge. Further comparisons with real wastewater comprising higher concentrations of lipids will be required to confirm whether the synergistic protein-lipid fouling behaviour observed in the model foulant analyses exists in real wastewater.

It was postulated here that particulate substrate will play a major role in industrial AnMBR bulk filtration. However, the identification of long term fouling layer composition in industrial AnMBRs is also important, i.e. whether substrate fouling will dominate, or whether the fouling layer will be affected by biomass concentration and the presence of soluble microbial products and extracellular polymeric substances. Membrane fouling layers with considerable microbial biomass, will not only affect filtration behaviour but can potentially have significant contributions to overall biological treatment capacity of the AnMBR [3].

6.3 OBJECTIVE 3: FOULING MODEL DEVELOPMENT AND MODEL BASED ANALYSES (CHAPTERS 4 AND 5)

Conclusions:
The effect of non-uniform shear was important in membrane fouling control in flat sheet membranes. At least 80% sparger coverage of membrane was required for adequate membrane shearing of the flat sheet AnMBR configuration. The model could be applied against experiments to quantify complex fouling parameters that cannot easily be measured in-situ, such as fibre induced membrane shear. Model estimated fibre shear enhancement factor, $\omega$, was approximately 5 times CFD shear in dilute wastewater (2 gTS/L). Shear enhancement was not identified in 35 gTS/L ($\omega = 1$) suggesting fibre movement is constrained in concentrated sludge. The model identified an optimum gas sparge rate of 10 L/min in total solids concentrations of 2 g/L to 20 g/L. Longer filtration periods at lower flux is a more beneficial strategy than shorter filtration periods and higher flux. Model predictions identified a linear relationship between required membrane surface area and sparger gas volume per day; and tank volume; with approximately 5 hollow fibre membrane modules and 76 m$^3$/day of gas required per m$^3$ tank volume.
Significance and implications of findings:
The development of a model based optimisation tool that combines the various interacting factors of membrane fouling and takes into consideration complex reactor hydrodynamics is an important contribution of this project to membrane bioreactor research. Application of a multidimensional CFD estimated shear into an overall membrane model enables the visualisation of planar cake and flux profiles in flat sheet membranes. Furthermore, the incorporation of a dynamic flux-pressure feedback loop in the model allows for the simulation of flux-step experiments and the prediction of critical flux. The model can be applied in the study of the effect of key design and operational parameters such as local geometry around the membrane, sparged gas flow rate, operating flux and the introduction of relaxation periods (gas sparging at no flux), on membrane performance and for the identification of their optimum values for the maximisation of membrane life. The model also allows for quantification of complex fouling parameters that cannot easily be measured in-situ, such as cake compressibility and complex fibre shear in hollow fibre membranes. This model will also be highly useful in a cost benefit analysis as it allows for dynamic and realistic operational analysis, and can be evaluated in long-term simulations to identify longer term strategies (under a dynamic regime).

Recommendations for future study:
Due to the capability of the model to predict cake accumulation, an important extension to the model would be to incorporate the potential biological treatment capacity of the fouling layer. This requires identification of biological kinetics within the fouling layer.

Future work to increase model applicability for fibre based membranes can include comprehensive filtration tests at a range of intermediate TS concentrations, fibre characteristics (packing density, length, diameter, tightness etc.) and sparge rates. Model based calibrations against these studies will quantify fibre-induced shear enhancement at each of these varying conditions; and can be correlated into an overall model using techniques such as Response Surface Methodology (RSM) that can be used to understand the combined effect of the various controlling factors in fibre-induced shear.

6.4 BROADER MODEL APPLICABILITY
The use of CFD allows model based fouling analysis in varying reactor designs and configurations. The membrane fouling model presented in this thesis with an updated CFD based estimation of membrane shear can be used to analyse the effect of non-uniform shear (Section 4.4.3) and hence also predict the effect of various sparger configurations and sparger flow rates. CFD modelling
importantly, allows for the investigation of scaling-up effects on membrane shear and fouling. The incorporation of CFD into the fouling model allows its use for design and optimisation of scaled-up geometry (tank width to depth ratio, position and number of sparger units for mixing and shearing, position and number of membrane modules, geometry and position of baffles, downcomer and riser zones near the membrane to enhance fluid velocity and shearing etc.), and scaled-up operational parameters (volume of sparger gas, optimum mixed liquor total solids etc.). The outcomes of lab and pilot-scale AnMBR filtration analyses can be made applicable to full-scale AnMBR geometries by updating the geometry in the CFD model and obtaining a shear profile. This shear profile can then be incorporated into the membrane fouling model in Chapter 4 to predict filtration behaviour in full-scale (assuming similar WW characteristics in lab-scale) and any differences in filtration behaviour between lab and full-scale due to hydrodynamics can be isolated and analysed. Full-scale reactor, membrane and sparger designs can be further optimised to maximise membrane shear and hence minimise membrane fouling.

The model was developed for submerged membrane systems, but can easily be applied in external membrane systems and in configurations where hydrodynamic shearing is used as fouling control. The CFD model can also simulate hydrodynamics in non-bubbled sparging systems which use particles (such as powdered activated carbon) for membrane shearing. The overall CFD modelling procedure will be the same, i.e. two phase with the dispersed phase characteristics to match those of the shearing particle.

The use of an incomplete mixing strategy that maintains higher sludge concentrations below the membrane can also be optimised using the model to minimise energy for mixing and membrane shearing.

The model will be further applicable in identification of short term filtration characteristics and cake fouling in aerobic MBRs as the primary fouling kinetics (convective flux and liquid/gas shearing) will be identical to anaerobic MBRs. The model does not include long-term, transient effects of the release of biological products (soluble microbial products, SMP and extracellular polymeric substances, EPS) and biofilm growth, which can play a major role in aerobic MBR fouling. However the model can be coupled with external SMP/EPS [4] and biofilm models to resolve these effects. Application of the model also extends to other industrial filtration processes, for example, those used in food and dairy processing.
6.5 REFERENCES

Figure A-1: Particle size distributions; a) individual components; b) individual and binary protein/carbohydrate mixtures; c) individual and binary protein/lipid mixtures; d) individual and binary carbohydrate/lipid mixtures and e) individual and ternary protein, carbohydrate and lipid mixtures.
Figure A-2: Contact angle in virgin (unused) membrane, 0.155 g protein fouled membrane, 1.0 g protein fouled membrane and 1.0 g lipid fouled membrane.