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Fouling in silicon microchannel designs used for IC chip cooling and its mitigation

Jeffrey L. Perry

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FOULING IN SILICON MICROCHANNEL DESIGNS USED FOR IC CHIP COOLING AND ITS MITIGATION

by

JEFFREY L. PERRY

A DISSERTATION

Submitted in partial fulfillment of the requirements for the degree of

Doctor of Philosophy

in

MICROSYSTEMS ENGINEERING

at the
Rochester Institute of Technology

October 2007

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Microsystems Engineering Program

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Approved by: ____________________________________________________________

Mustafa A.G. Abushagur
Director of Microsystems Engineering Program

Certified by: _____________________________________________________________

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Dean Kate Gleason College of Engineering
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Fouling in Silicon Microchannel Designs Used for IC Chip Cooling and Its Mitigation

By

Jeffrey L. Perry

Submitted by Jeffrey L. Perry in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Microsystems Engineering and accepted on behalf of the Rochester Institute of Technology by the dissertation committee.

We, the undersigned members of the faculty of the Rochester Institute of Technology, certify that we have advised and/or supervised the candidate on the work described in this dissertation. We further certify that we have reviewed the dissertation manuscript and approve it in partial fulfillment of the requirements of the degree of Doctor of Philosophy in Microsystems Engineering.

Approved by:

Dr. Satish G. Kandlikar (Committee Chair and Dissertation Advisor) Date

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Dr. Michael A. Jackson

Dr. James E. Moon

MICROSYSTEMS ENGINEERING PROGRAM
ROCHESTER INSTITUTE OF TECHNOLOGY
October 2007
Particulate fouling studies with alumina and silica dispersions were performed in silicon, rectangular microchannels having hydraulic diameters between 220-225 µm. The particulates used ranged from the colloidal size range up to tens of microns in size (for particle aggregates). Data show for the most part the absence of particle depositions within the microchannels. This is even the case when there is an electrostatic attraction between the particles and the microchannel surface. The primary reason for this is due to the high wall shear stress at the microchannel walls.

In contrast, the headers for the microchannels are quite susceptible to particulate fouling under the same conditions. This is because the shear stress at the surface is lower. This fouling within the header, however, does not provide an increase in pressure drop within the microchannel device. Moreover, depositions within the header region can be mitigated with proper pH adjustment.

There is a secondary effect in particulate fouling when fibrous elements exist within the particle dispersion. The fouling behavior due to fibrous material is quite different. In fact, the presence of fibers is extremely detrimental to pressure drops within a microchannel device.

A multi-scale force balance model for particulate fouling is developed. It uses conventional theories on the forces between a particle and a wall which are extended to particulate fouling within a microchannel device. The model covers a scale spanning several orders of magnitude. In addition, it includes van der Waals forces, electrostatic forces, fluid-mechanics-related forces due to shear/lift, and a body force due to gravity.
ACKNOWLEDGMENTS

I would like to express my gratitude to Dr. Satish Kandlikar for taking me on as his graduate student and for his guidance throughout this project. I am grateful for Dr. Santosh Kurinec, Dr. Michael Jackson and Dr. James Moon for being on my thesis committee and being willing to donate time to answer my questions. I acknowledge Dr. Matthew Yates at the University of Rochester for generous use of his instrumentation for particle size analysis and zeta potential measurements. I also appreciate the help of Brian McIntyre at the University of Rochester for performing Scanning Electron Microscope analyses on my particles. I give my sincere appreciation to my father, James Perry, and wife, Josephine Perry, for providing help in numerous ways, including financial and emotional support for my many years at RIT. In addition, I am appreciative of my son, Jeremiah Perry, who helped inspire me to finish my dissertation work in order to support the family. I hope my education at RIT will be able to provide for him in all the coming years.
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a  Particle radius (m) or a constant or channel side (m)
A_{132} Hamaker constant (J)
A_c Channel cross-sectional area (m^2)
b  Half channel width (m) or a constant or channel side (m)
c  Half channel height (m) or a constant
C_b Bulk concentration (kg/m^3)
C_w Concentration of particles adjacent to surface (kg/m^3)
D  Diameter of pipe (m)
D_B Brownian diffusivity (m/s^2)
D_h Hydraulic diameter (m)
d_p Particle diameter (m)
e  Elementary electric charge (coulomb)
E  Interaction energy (J)
E_{el} Electrostatic interaction energy (J)
E_t Total interaction energy (J)
E_{vdw} Van der Waals interaction energy (J)
f  Fanning friction factor (dimensionless)
f_{app} Apparent friction factor (dimensionless)
F_D Drag force (N)
F_{el} Electrostatic force (N)
F_g Gravitational force (N)
F_L Lift force (N)
F_o f-test value (dimensionless)
F_T Total force (N)
F_{vdw} Van der Waals force (N)
g  Acceleration of gravity (m/s^2)
G  Ratio of V_G to U_\infty (dimensionless)
h  Surface roughness (m)
H Minimum separation distance (m)
J Value used in determining \eta_{imp} (dimensionless)
k Boltzmann constant (J/K)
<table>
<thead>
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<tr>
<td>$k_m$</td>
<td>Mass transport coefficient (m/s)</td>
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<td>$k_m^+$</td>
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<td>$k_{hf}$</td>
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<tr>
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<td>$m$</td>
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<td>Y</td>
<td>Distance of particle from centerline of fiber (m)</td>
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<tr>
<td>z</td>
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**Greek**

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<td>α</td>
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<td>Asymptotic value in thermal fouling (1/s) or Angle (degree)</td>
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<td>Stern layer thickness (m)</td>
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<td>Parameter in determining zeta potential (dimensionless)</td>
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<td>( \varepsilon )</td>
<td>Permittivity of medium (F/m)</td>
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<td>( \varepsilon_r )</td>
<td>Relative dielectric constant of medium (dimensionless)</td>
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<td>Deposit mass flowrate per unit area (kg/s·m(^2))</td>
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<tr>
<td>( \phi_R )</td>
<td>Removal mass flowrate per unit area (kg/s·m(^2))</td>
</tr>
<tr>
<td>( \eta_{\text{diff}} )</td>
<td>Single-fiber efficiency due to diffusion (dimensionless)</td>
</tr>
<tr>
<td>( \eta_{\text{grav}} )</td>
<td>Single-fiber efficiency due to gravity (dimensionless)</td>
</tr>
<tr>
<td>( \eta_{\text{imp}} )</td>
<td>Single-fiber efficiency due to inertial impact (dimensionless)</td>
</tr>
<tr>
<td>( \eta_{\text{int}} )</td>
<td>Single-fiber efficiency due to interception (dimensionless)</td>
</tr>
<tr>
<td>( \eta_{\text{total}} )</td>
<td>Total single-fiber efficiency due to all mechanisms (dimensionless)</td>
</tr>
<tr>
<td>( \kappa )</td>
<td>Debye-Hückel parameter (1/m)</td>
</tr>
<tr>
<td>( 1/\kappa )</td>
<td>Debye length or diffuse layer thickness (m)</td>
</tr>
<tr>
<td>( \lambda )</td>
<td>Wavelength (m)</td>
</tr>
<tr>
<td>( \lambda_c )</td>
<td>Characteristic wavelength of interaction (m)</td>
</tr>
<tr>
<td>( \lambda_\text{e} )</td>
<td>Emitted wavelength (m)</td>
</tr>
<tr>
<td>( \mu )</td>
<td>Mean value (units can vary)</td>
</tr>
<tr>
<td>( \mu_E )</td>
<td>Electrophoretic mobility (m(^2)/V·s)</td>
</tr>
<tr>
<td>( \mu_f )</td>
<td>Viscosity of fluid (kg/m·s)</td>
</tr>
<tr>
<td>( \nu )</td>
<td>Kinematic viscosity (m/s(^2))</td>
</tr>
<tr>
<td>( \theta )</td>
<td>Angle (degrees)</td>
</tr>
<tr>
<td>( \rho_f )</td>
<td>Density of fluid (kg/m(^3))</td>
</tr>
<tr>
<td>( \rho_p )</td>
<td>Density of particle (kg/m(^3))</td>
</tr>
<tr>
<td>( \rho_{\text{fr}} )</td>
<td>Density of foulant (kg/m(^3))</td>
</tr>
<tr>
<td>( \sigma )</td>
<td>Standard deviation (units can vary)</td>
</tr>
<tr>
<td>( \tau_w )</td>
<td>Shear stress at wall (Pa)</td>
</tr>
<tr>
<td>( \psi )</td>
<td>Steam function (m(^2)/s)</td>
</tr>
<tr>
<td>( \psi_d )</td>
<td>Stern potential (V)</td>
</tr>
<tr>
<td>( \psi_s )</td>
<td>Surface potential (V)</td>
</tr>
<tr>
<td>( \zeta )</td>
<td>Zeta potential (V)</td>
</tr>
</tbody>
</table>
### LIST OF ABBREVIATIONS

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>APS</td>
<td>Approximate Particle Size</td>
</tr>
<tr>
<td>BFSL</td>
<td>Best Fit Straight Line</td>
</tr>
<tr>
<td>CAD</td>
<td>Computer Aided Design</td>
</tr>
<tr>
<td>CCD</td>
<td>Charged Coupled Device</td>
</tr>
<tr>
<td>CFI</td>
<td>Chromatic Aberration-Free Infinity</td>
</tr>
<tr>
<td>DAQ</td>
<td>Data Acquisition</td>
</tr>
<tr>
<td>DLVO</td>
<td>Derjaguin Landau Verwey Overbeek</td>
</tr>
<tr>
<td>DRIE</td>
<td>Deep Reactive Ion Etching</td>
</tr>
<tr>
<td>FE</td>
<td>Field Emission</td>
</tr>
<tr>
<td>FS</td>
<td>Full Scale</td>
</tr>
<tr>
<td>IC</td>
<td>Integrated Circuit</td>
</tr>
<tr>
<td>ITRS</td>
<td>International Technology Roadmap for Semiconductors</td>
</tr>
<tr>
<td>MEMS</td>
<td>Micro Electrical Mechanical Systems</td>
</tr>
<tr>
<td>NIST</td>
<td>National Institute of Standards and Technology</td>
</tr>
<tr>
<td>OSTLE</td>
<td>One Step Two Level Etching</td>
</tr>
<tr>
<td>PIV</td>
<td>Particle Image Velocimetry</td>
</tr>
<tr>
<td>PPM</td>
<td>Parts Per Million</td>
</tr>
<tr>
<td>PTFE</td>
<td>Poly Tetra Fluoro Ethylene</td>
</tr>
<tr>
<td>PZC</td>
<td>Point of Zero Charge</td>
</tr>
<tr>
<td>ROI</td>
<td>Region of Interest</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscope</td>
</tr>
<tr>
<td>YAG</td>
<td>Yttrium Aluminum Garnet</td>
</tr>
</tbody>
</table>
CHAPTER 1

INTRODUCTION

Microfluidics is an increasingly studied field because of its use in practical applications in both biology and microelectronics. In recent years, the proliferation of MEMS (Micro Electro Mechanical Systems) has resulted in the use of microchannels for many applications. In particular, with microchannels there has been a great emphasis on semiconductor chip cooling. Consequently, it is desirable to study the effect of particulates in such devices which must operate over the lifetime of the semiconductor chip.

1.1 Motivation

This thesis is being conducted to understand particulate fouling in microchannels which is largely unexplored. As part of this endeavor, it is desired to conduct fundamental experiments to gain badly needed information on this phenomenon. The main premise of this work is that there is a need to explore this area because in microchannel fouling the hydraulic diameters are very small and, consequently, microchannels may be susceptible to excessively large pressure drops. Moreover, the utility and lifetime of certain microfluidic devices depend critically on their ability to maintain flow without interruption as with a microchannel heat exchanger used for semiconductor chip cooling. Because of their small size, microchannels can be blocked by small amounts of foulant matter. In the present context particulate fouling is defined as the accumulation of solid particles suspended in a fluid onto the microchannel surface.
As microchannel heat exchangers enter the market place and experience extensive use for commercial applications the importance of particle adhesion and clogging will eventually limit their reliability for long term use. This reality is further underscored by the fact that heat dissipation requirements continually increase for semiconductor chip cooling because of the increased number of transistors in a semiconductor chip. This is governed by Moore’s law which states that the number of transistors that can fit on a single computer chip doubles approximately every 18 months. According to the International Technology Roadmap for Semiconductors (ITRS) the maximum power requirements for a single chip package will reach 108 W/cm² by 2016. Figure 1.1 shows the prediction from the 2006 ITRS report. Consequently, fouling within microchannel heat exchangers must be understood and mitigated before the technology can be guaranteed to deliver the required heat dissipation if the technology trend is expected to continue.

![Figure 1.1. Maximum heat dissipation requirements for a single chip package. Data from ITRS 2006.](image-url)
Another aspect that has direct application to particulate fouling is the use of nano-fluids in microchannel heat exchangers for improved heat management. Heat transfer by convection depends in part on the thermal conductivity of the fluid. Therefore, superior performance is achieved by increasing the thermal conductivity of the fluid. This is done by using a suspension of solid particles in a fluid for improved heat transfer. This is possible since the thermal conductivity of solids is generally higher than the fluid. Studies of nano-fluids have shown substantial increases in the thermal conductivity of fluids by adding very small amounts of metallic or metallic oxide nano-particles (Cu, CuO, Al₂O₃) to the fluid. Vadasz et al. (2005). Lee et al. (1999), reported that the addition of 4.3 vol.% Al₂O₃ nano-particles to water increased the thermal conductivity by 30%.

A major drawback to the technology is the abrasive action of particles which can cause the erosion of components and clogging because the flow passages are very small, resulting in prohibitively large pressure drops. In addition, because of the small size of the nano-particles, there is an increased tendency for particle aggregation which results from the lowered force of repulsion between them. This can have severe consequences in the overall performance. This further emphasizes the need for fouling studies to be conducted in microchannel devices.

Both aspects which were previously discussed in this section can also benefit from a basic model to help predict when particulate fouling will occur within a microchannel device. This will help understand the fouling phenomenon better so that methods can be developed to mitigate its detrimental effects.
1.2 Previous Research

The papers discussed in this section are shown in Table 1.1. The smallest hydraulic diameters, $D_h$, that have been previously used in fouling studies that the author is aware of were by Benzinger et al. (2005). Their microstructured heat exchanger had a $D_h = 178 \, \mu m$. Their studies focused on crystallization fouling using aqueous solutions of $\text{Ca(NO}_3\text{)}_2/\text{NaHCO}_3$. However, crystallization fouling is an unlikely occurrence in microchannels used for heat dissipation as it relates to semiconductor chip cooling.

Concerning particulate depositions, however, the smallest hydraulic diameters used were in the studies of Yiantsios et al. (1995, 1998, 2003) and Niida et al. (1989). Yiantsios et al. studied fouling in a single channel with $D_h = 952 \, \mu m$. Their test section consisted of two glass plates separated by Teflon spacer strips to create a channel that was 10 mm by 0.5 mm. Similarity, Niida et al. used an observation cell, made of borosilicate glass with a cross-section of 4 mm by 0.4 mm to provide a $D_h$ of 727 $\mu m$. In the present fouling studies microchannels are being used which have dimensions of 249 $\mu m \times 205 \, \mu m$ ($D_h = 225 \, \mu m$) and 407 $\mu m \times 151 \, \mu m$ ($D_h = 220 \, \mu m$).

Additional work has been done by Sharp et al. (2001, 2005) who looked at flow-blocking particle structures in microtubes with diameters between 75 and 406 $\mu m$. They, however, only investigated conditions when the particle diameter to microtube diameter ($d_p/D$) was rather large, $d_p/D = 0.24 – 0.49$. In this regime flow obstruction was caused solely by shear induced arching. This occurs when particle diameters are of the order of the flow passage. In this case arching refers to situations in which particles can form an arch structure that is capable of supporting a load through simple contact stresses.
between the particles. A stable arch structure is able to form within the channel and cause flow blockage.

In general particulate fouling may be considered a two-step process. It consists of a transport step, in which particles are transferred to the channel wall, and a subsequent adhesion step, which is dominated by the interaction forces between particles and the wall. An important parameter in both steps is the particle size. It is a critical parameter that determines the particle attachment efficiency. In fact, the particle size affects the magnitude of physicochemical interactions between particles and a channel wall as well as the hydrodynamic forces that tend to cause particles to detach or prevent them from adhering.

### Table 1.1. Papers whose research was on fouling or flow blocking within microchannels.

<table>
<thead>
<tr>
<th>Author</th>
<th>Year</th>
<th>$D_h$ (µm)</th>
<th>Research</th>
</tr>
</thead>
<tbody>
<tr>
<td>Niida et al.</td>
<td>1989</td>
<td>727</td>
<td>Particle deposits within a single microchannel</td>
</tr>
<tr>
<td>Yiantsios et al.</td>
<td>1995, 1998, 2003</td>
<td>952</td>
<td>Particle deposits within a single microchannel</td>
</tr>
<tr>
<td>Sharp et al.</td>
<td>2001, 2005</td>
<td>75-406</td>
<td>Flow blocking</td>
</tr>
<tr>
<td>Benzinger et al.</td>
<td>2005</td>
<td>178</td>
<td>Crystallization fouling</td>
</tr>
</tbody>
</table>

### 1.3 Objectives

The overriding scope of the present work is to provide engineers and scientist working with microchannels the ability to mitigate fouling in microchannel devices
which can limit device functionality. Understanding in the fundamental aspects of microchannel fouling is necessary before they can be utilized on a large scale for commercial applications. To this end, the location of where an increase in pressure drop due to particulate fouling might occur in a microfluidic device needs to be determined. It is possible that fouling in a microchannel device may occur at (1) the inlet and outlet headers of the microchannels where fluid velocities may not be great enough to help prevent particulates from depositing, (2) bends in micro-passages where particulates may accumulate due to geometrical effects or flow separation, and (3) within the microchannels themselves which have the smallest geometrical constraints.

There are several major objectives of the present work. They are as follows:

- Understand the nature of particulate fouling in microchannels.
- Identify which parameters can most effectively mitigate particulate fouling within a microchannel device.
- Develop a force-based model in particulate fouling of microchannels.
- Develop an experimental setup and procedures to study particulate fouling in microchannels.
- Determine where particulate depositions are most likely to occur in a microchannel device and what effect they will have on pressure drops.
- Propose microchannel configurations that will mitigate particulate fouling.
- Reveal what aspects of fouling in typical microchannel devices are most detrimental for increased pressure drops.
CHAPTER 2

LITERATURE REVIEW

The term “fouling” was originally used in the oil industry and became widely used in literature to describe any undesirable deposit which caused an increase in the thermal resistance of a heat exchanger, Liu (1999). In addition to the increase in thermal resistance, fouling can reduce the hydraulic performance which can lead to excessive pressure drops. All of the literature available in fouling is nearly exclusive to studying the increase in thermal resistance; however, attempts will be made to draw inferences on how these basic fundamentals can be used to understand fouling as it relates to an increase in differential pressure within a microchannel device.

2.1 Types of Fouling

Fouling of a heat exchanger can be the result of one or more of a number of mechanisms. It is generally accepted that the fouling processes can be categorized into one of seven groups. Each one will be briefly discussed here.

1. **Particulate Fouling**: This is the accumulation of finely suspended solid particles onto the heat transfer surface. The particles are usually small in size with diameters less than several microns. Particulate fouling does not usually result in thick deposits, and the particles are easily removed unless accompanied by some other mechanism which helps “cement” them to the surface.

2. **Crystallization Fouling or Scale Formation**: Deposits of this type occur when salts are dissolved in the process fluid which crystallizes onto the heat transfer
surface because the solubility of the dissolved ions changes with temperature. In order for scale formation to occur supersaturation of the depositing species in solution must be present near the solid surface. Crystallization depositions can become very thick and in some instances blocked tubes have been reported, Bott (1990).

3. **Chemical Reaction Fouling**: Deposit formation occurs at the heat transfer surface by chemical reactions in which the surface material itself is not a reactant. Reaction fouling can occur over a wide range of temperature from ambient up to those found in combustion systems. However, it is usually more pronounced at higher temperatures, Bott (1990).

4. **Corrosion Fouling**: This involves the accumulation of deposits which are formed with the assistance of the heat transfer surface material which has been attacked chemically by the process fluid. It is the presence of corrosion products on the surface that constitutes a fouling deposit.

5. **Biological Fouling**: This includes the attachment and subsequent growth of both macro and micro-organisms onto the heat transfer surface. Macrofouling is usually associated with larger species such as mussels, barnacles and vegetation while microfouling occurs from micro-organisms such as bacteria, fungi and algae. Since this type of fouling involves living matter, the temperature range over which it can exist is limited. Microfouling, typically occurs between 0 and 90°C, Bott (1990).

6. **Solidification Fouling**: This is the freezing of a pure liquid or one component from a liquid phase onto a subcooled surface.
7. **Mixed Fouling**: This is the presence of two or more of the above fouling mechanisms which occur simultaneously.

2.2 **Characterization of Fouling in Smooth Channels**

As mentioned at the beginning of this chapter, the figure of merit in fouling has almost exclusively been the increase in thermal resistance. Although reduced heat transfer efficiency is of prime importance even with a microchannel heat exchanger, it can be argued that studying the pressure drops in this type of microchannel device are of greater practical interest. This is because the utility and lifetime of such a device depends critically on its ability to maintain flow without interruption which can be blocked by even small amounts of matter.

This study uses the pressure drops across a microchannel device as the figure of merit in fouling. However, because the literature available is based on thermal resistance, this aspect will be touched on so that inferences can be made. The thermal resistance, \( R_t \), in conductive heat transfer is given by Eq. 2.1

\[
R_t = \frac{T_{s,1} - T_{s,2}}{q} = \frac{\Delta T}{q}
\]  

(2.1)

where \( \Delta T \) is temperature difference between two surfaces of temperature \( T_{s,1} \) and \( T_{s,2} \) with a heat flux \( q \) flowing between them. Figure 2.1 helps depict this. In fouling the particle deposition onto a heat conducting surface will create a layer of unwanted material which will have a lower thermal conductivity than the heat transfer surface. This in turn creates a greater resistance to heat transfer. In thermal fouling, the fouling rate can be calculated from following expression
where the change in thermal resistance is calculated at some point in time. The subscripts \( t \) and \( 0 \) represent time at, time = \( t \) and time = \( 0 \), respectively. From Eq. 2.2 it becomes obvious that fouling is a time-dependent phenomena.

Kern and Seaton (1959) were the first to offer a general expression for predicting the rate of build-up for the amount of deposit with mass, \( m \), over a time, \( t \). Their expression was inspired by their observation of refinery heat exchangers which had an asymptotic type deposition curve. They suggested a fouling model based on a mass balance where the net fouling rate is the difference between the solid’s deposition rate and its removal rate. This can be stated mathematically as

\[
\frac{dM}{dt} = \phi_D - \phi_R
\]

where \( M \) is the mass deposit per area and \( \phi_D \) and \( \phi_R \) are the deposit and removal mass flowrates per unit area of surface, respectively.
Figure 2.2 shows three idealized fouling curves. The first curve (A) depicts a linear growth of the fouling deposit which occurs if $\phi_D$ and $\phi_R$ are constant or if $\phi_R$ is negligible. The second curve (B) illustrates a falling rate deposition where there is a steady decrease in the difference between $\phi_D$ and $\phi_R$. This difference approaches zero as $R_f$ approaches infinity. The third type of fouling curve (C) commonly observed is where the fouling resistance attains an asymptotic value. This occurs when there is a steadily decreasing difference between $\phi_D$ and $\phi_R$, in which the difference approaches zero as $R_f$ approaches a finite value. It should be mentioned that unless the fouling process has been carried out for a sufficiently long period of time, one cannot be sure whether or not an apparent linear curve will eventually exhibit falling rate behavior or if a falling rate curve will ultimately behave asymptotically.

Figure 2.2. Idealized fouling curves.
The curves in Figure 2.2 show a portion of time where there is little evidence of fouling behavior. This is referred to as the delay or induction period where the heat transfer surface is modified in some way so that fouling eventually takes place. It is often extremely difficult or impossible to predict the initiation period even with the benefit of experience. Therefore, most mathematical models that have been developed in fouling ignore it.

The curves represented in Figure 2.2 are ideal. In a more practical situation this ideality may not be achieved. A more realistic asymptotic curve is given in Figure 2.3 where a “saw tooth” effect is observed from the partial removal of deposits due to spalling, to be followed for a short time by a rapid build up of material. Similar effects of partial removal and deposition may also be observed with the other fouling curves.

Figure 2.3. Saw tooth fouling curve. A smooth curve is approximated with the dotted line.
2.2.1 Liner Model

The simplest model is curve A of Figure 2.2. If the induction period is ignored, as is most often done, it will have the form given in Eq. 2.4

\[ R_{tf} = \frac{dR_{tf}}{dt} t \]  

(2.4)

where \( R_{tf} \) is the thermal fouling resistance at time \( t \) and the differential \( dR_{tf}/dt \) is just the slope of the line. Liner fouling behavior has been associated with the crystallization of well-formed deposits which have not been contaminated by the presence of co-precipitated impurities. The strong bonds that characterize this type of fouling prevent significant removal rates and hence give linear fouling, Zubair et al (2000).

2.2.2 Falling Rate Model

The falling rate model is illustrated in curve B of Figure 2.2. As the name implies it is neither linear nor asymptotic in nature. It therefore, has no general mathematical model which can readily be applied to describe its behavior. Consequently, it has a somewhat elusive quality. As mentioned earlier this may be due to the fact that the fouling process has not been carried out long enough to reach an asymptotic value. All that can readily be inferred about this type of process is that there exists a steady decrease in the difference between the deposition and removal rates over time as given by Eq. 2.3.

2.2.3 Asymptotic Model

Fouling curve C of Figure 2.2 is discussed here. It is described mathematically in Eq. 2.5 which was put forth originally by Kern and Seaton, (1959)
\[ R_f = R_{f\infty} (1 - e^{-\beta t}) \]  

where \( R_{f\infty} \) is the thermal fouling resistance at infinite time – the asymptotic value, and \( \beta \) is a constant which is dependent on the system properties and operating conditions. Using the work of Kern and Seaton it is possible to study the mass deposits of a foulant by modifying the previous equation to give the following

\[ M = M^* (1 - e^{-\beta t}) \]  

where \( M \) is the mass deposit per unit area at time \( t \) with \( M^* \) as its asymptotic value. The relationship between Eqs. 2.5 and 2.6 is given by the following expression at a given time

\[ R_f = \frac{M}{\rho_{tf} k_{tf}} \]  

where \( \rho_{tf} \) and \( k_{tf} \) are the density and thermal conductivity of the foulant, respectively. The work of Kern and Seaton gives \( M^* \) and \( \beta \) as

\[ M^* = \frac{a_1 C_b V_m}{a_2 \tau_w} \]  

\[ \beta = a_2 \tau_w \]  

where \( a_1 \) and \( a_2 \) are constants, \( C_b \) is the bulk concentration of the fouling material, \( V_m \) is the mean fluid velocity and \( \tau_w \) is the wall shear stress. Kern and Seaton derived their expression in Eq. 2.5 by integrating Eq. 2.3 where \( \phi_D \) and \( \phi_R \) were defined based upon observation and experience in terms of process parameters. The corresponding mass fluxes used in their modeling are given below.

\[ \phi_D = a_1 C_b V_m \]  

\[ \phi_R = a_2 \tau_w M \]
2.3 Particulate Fouling

Particulate fouling may be considered a three-step process. It first consists of a transport step, in which particles are transferred to the channel wall from the bulk solution. The second step involves the adhesion of the particles to the surface under favorable conditions. Finally the third step consists of detachment, which occurs if the removal forces are sufficiently strong to overcome the adhesion forces.

2.3.1 Particle Transport

In general the arrival of a particle towards a surface can be attributable to one of two mechanisms. They are gravitational settling and particle transport within the fluid as it moves across the surface. In most practical heat exchangers, gravity does not play any significant role and is therefore generally ignored.

A concentration gradient is necessary to drive a mass transfer process. Figure 2.4 shows a particle concentration profile for a fluid passing by a surface.

![Figure 2.4. Particle concentration profile for a fluid passing by a surface.](image)
The depositional mass flux \( \phi_D \) is defined as

\[
\phi_D = k_m (C_b - C_w)
\]  

(2.12)

where \( k_m \) is the mass transport coefficient, \( C_b \) is the bulk concentration of the particles and \( C_w \) is the concentration of the particles adjacent to the wall. If the particles adhere completely to the wall, then \( C_w = 0 \). In this case

\[
\phi_D = k_m C_b
\]  

(2.13)

It is possible for some of the particles to hit the surface and then rebound off of it. In this situation the term sticking probability is used. As the name implies it is an attempt to quantify the retention of the particles on the surface. Using this modification Eq. 2.13 becomes

\[
\phi_D = k_m C_b P
\]  

(2.14)

where \( P \) is the sticking probability. The sticking probability will have a value less than unity. Its magnitude will depend on the conditions associated with the flowing fluid, the size and nature of the particles and the physicochemical interactions between the particles and the walls of the conduit. The sticking probability is very difficult to determine and it is likely to change with time as the deposition continues.

**2.3.2 Particle Transport – Isothermal Conditions**

Much of the theory developed for the transport of particles from a bulk solution to a nearby surface has been under isothermal conditions. Although this may seem somewhat limiting theoretically, the experiments conducted within this body of work are themselves isothermal. Figure 2.5 shows a particle approaching a surface. It is assumed that the particle makes its approach at a right-angle to the surface from a distance equal to
the stopping distance. Furthermore, it is also implicitly understood that the particle has received sufficient momentum from turbulent eddies to cross the viscous sublayer by its inertia and will eventually come to rest on the surface.

Figure 2.5. Idealized approach of a particle through the viscous sublayer towards a surface due to turbulent eddies.

The stopping distance, $S$, of a particle is defined as the distance a particle will travel though a stationary fluid when subjected to some initial velocity $V_o$. The relation between these two quantities is

$$S = t_r V_o$$  \hspace{1cm} (2.15)

where $t_r$ is the particle relaxation time. $t_r$ is for particles subject to the aerodynamic resistance of Stokes drag which is given by

$$t_r = \frac{\rho_p d_p}{18 \mu_f}$$  \hspace{1cm} (2.16)
where $\rho_p$ is the particle density, $d_p$ is the particle diameter and $\mu_f$ is the fluid dynamic viscosity. A dimensionless particle relaxation time $t_r^+$ is obtained by multiplying by $(V^*)^2/\nu$ as shown in Eq. 2.17.

$$t_r^+ = \frac{t_r (V^*)^2}{\nu}$$

with $V^*$ as the friction velocity and $\nu$ as the kinematic viscosity which is given by

$$V^* = \sqrt{\frac{\tau_w}{\rho_f}} = V_m \sqrt{\frac{f}{2}}$$

where $f$ is the fanning friction factor.

It is possible to develop a dimensionless mass transfer coefficient $k_m^+$ if $k_m$ is divided by $V^*$ as shown in Eq. 2.19

$$k_m^+ = \frac{k_m}{V^*}$$

When $k_m^+$ is plotted against $t_r^+$ three separate transport regimes are exhibited. This is clearly demonstrated in Figure 2.6 which comes from a review paper by Papavergos and Hedley (1984). Papavergos and Hedley made the useful demarcations in regimes for isothermal particle deposition which are given in Table 2.1. Only the diffusion regime has application to particle deposition inside microchannels because the flow is laminar. However, for completeness all three will be discussed.

<table>
<thead>
<tr>
<th>$t_r^+$</th>
<th>Regime</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt; 0.2$</td>
<td>Diffusion</td>
</tr>
<tr>
<td>0.2 - 20</td>
<td>Inertia</td>
</tr>
<tr>
<td>$&gt; 20$</td>
<td>Impaction</td>
</tr>
</tbody>
</table>
Figure 2.6. Dimensionless deposition velocity (or $k_m^+$) vs. particle relaxation time (given as $t_r^+$ in text), Papavergos and Hedley (1984).

2.3.2.1 Diffusion Particle Transport

Particles suspended in the fluid are carried towards the surface by Brownian motion. This will occur under laminar flow conditions or for turbulent flow once the particle has made it into the laminar sublayer. Under these circumstances the small particles involved are treated as molecules. In accordance with this, general mass transfer theory can be applied in calculating $k_m$. The Brownian diffusivity, $D_B$ may be calculated for a dilute suspension of spheres using the Stokes-Einstein equation

\[
D_B = \frac{kT}{3\pi\mu_f d_p} \tag{2.20}
\]
where $T$ is the absolute temperature of the suspension and $k$ is the Boltzmann constant. This equation shows that the Brownian diffusivity increases as particle size decreases. In addition, the deposition rate will increase at higher temperatures because of the temperature term in the numerator and due to the viscosity which decreases at higher temperatures. The mass transfer coefficient, $k_m$, can be estimated by the following equation

$$Sh = \frac{k_m D_h}{D_B} = a \text{Re}^b \text{Sc}^c$$

(2.21)

where the additional terms are defined below.

$Sh = \text{Sherwood number}$

$Re = \text{Reynolds number} = \frac{\rho_f V_m D_h}{\mu_f}$

$Sc = \text{Schmidt number} = \frac{\mu_f}{\rho_f D_B}$

$a = \text{constant}$

$b$ and $c$ are numerical indices and $D_h$ is the hydraulic diameter.

Masri and Cliffe (1996) used this type of correlation to study particle deposition inside a compact plate fin heat exchanger which utilized minichannels. The correlation was originally developed for laminar flow parallel to a flat plate\(^1\). The correlation used to estimate the particle mass transfer coefficient inside a channel was

$$Sh = 0.664 \text{Re}^{0.50} \text{Sc}^{0.33}$$

(2.22)

\(^1\) Actual correlation came from textbook by Eckert and Drake (1959).
2.3.2.2 Inertial Particle Transport

In the inertia regime the particles have obtained sufficient energy from the fluid to enable them to move through the viscous sublayer under turbulent flow. Particles on the low end of the regime (small $t_r^+$) will eventually slow down and finally be arrested by drag forces in the laminar sublayer and become subject to Brownian motion. Other particles at the high end of the regime (large $t_r^+$) possess sufficient energy in order to reach the surface. Papavergos and Hedley (1984) suggested as a rough empirical generalization the following equation

$$k_m^+ = 3.5 \times 10^{-4} (t_r^+)^2$$  \hspace{1cm} (2.23)

2.3.2.3 Impaction Particle Transport

In this regime the particle velocity that propels it towards the wall approaches the friction velocity $V^*$. Consequently, the particle stopping distance is on the same order as the hydraulic diameter. The particles in this regime have enough momentum that when they are exposed to turbulent fluctuations there is little effect. Papavergos and Hedley (1984) recommended the following approximation in determining $k_m^+$

$$k_m^+ = 0.18$$  \hspace{1cm} (2.24)

2.4 Particle Adhesion

In general, the phenomenon of fouling represents the interaction between solids. At the onset of the fouling process the interaction is between the heat exchanger surface and the foulant. However, as the deposit thickness increases the primary interaction is
involved between the foulant material that was previously deposited and new foulant particles which are arriving at the fluid/solid interface.

Adhesion is therefore a complex process as far as heat exchanger fouling is concerned, particularly where mixed fouling mechanisms are present or when different types of solid particulates exist in the same fluid. It is useful, however, to consider the underlying theory which influences this phenomenon of adhesion. To this end, adhesion will be considered in terms of a solid sphere approaching a flat plate.

### 2.4.1 Van der Waals Force between Particle/Wall

The van der Waals or London dispersion force is an attractive force between two atoms or non-polar molecules that arises due to fluctuating dipole moments. This interaction potential is largely insensitive to variations in electrolyte concentration and pH, and so may be considered as fixed in a first approximation, Israelachvili (1992). In this phenomenon a fluctuating dipole moment in one molecule induces a dipole moment in the other which results in a net attraction between them. With the exception of highly polar molecules, London dispersion forces account for nearly all of the van der Waals attraction which is operative. The van der Waals force between a sphere and a plate, which takes into account retardation effects, is given by Gregory (1981)

\[
F_{\text{vdw}} = \frac{A_{12}d_p}{12H^2} \left[1 - \frac{1}{1 + \frac{\lambda_c}{5.32H}}\right], \quad H \ll \frac{d_p}{2}
\]

---

2 There are three types of van der Waals forces, (1) those that exist between two permanent dipoles which orient each other such that on average attraction results, (2) when dipolar molecules induce dipoles in other non-polar molecules so that attraction occurs, and when (3) attraction occurs between non-polar molecules.
where $A_{132}$ is the Hamaker constant of particle 1 on wall 2 in medium 3, $d_p$ is the particle diameter, $H$ the minimum separation between the particle and the wall, and $\lambda_c$ is the characteristic wavelength of interaction which is taken as $\approx 100$ nm for most materials and compensates for the retardation effect, Gregory (1981). The Hamaker constant for single materials is usually between $10^{-19}$ and $10^{-20}$ J. Retardation occurs because it takes a finite amount of time for an electromagnetic signal to travel back and forth between two molecules. When the electromagnetic signal returns to the original emitting molecule the dipole moment will have changed direction and magnitude. This causes the dispersion force to be weakened or retarded. Eq. 2.25 is an adequate approximation for all separation distances up to about 5% of the particle diameter. This restriction is imposed by the use of the Derjaguin procedure; Hunter (2001), Hogg et al. (1965). The presence of a liquid dispersion medium, rather than a vacuum between the particle and the wall, notably lowers the van der Waals force. The effective Hamaker constant in his case for particle 1 near wall 2 in medium 3 is

$$A_{132} = A_{12} + A_{33} - A_{13} - A_{23} \quad (2.26)$$

If the attractions between unlike phases is taken to be the geometric mean of the attractions of each phase itself, $A_{12} = (A_{11} \times A_{22})^{1/2}$, then Eq. 2.26 becomes

$$A_{132} = \left(\sqrt{A_{11}} - \sqrt{A_{33}}\right)\left(\sqrt{A_{22}} - \sqrt{A_{33}}\right) \quad (2.27)$$

### 2.4.2 Electrostatic Force between Particle/Wall

Most materials immersed in an aqueous environment acquire a surface charge due to preferential adsorption of ions present in the liquid phase or due to dissociation of surface groups. Since a surface/water system must remain as a whole electrically neutral,
the dispersion medium must contain an equivalent charge of the opposite sign. This sets up the situation where there will be an excess of ions of one sign at the surface interface and an excess of ions of opposite sign in the solution near the surface. The electrical forces that are present because of the formation of this space charge are counteracted by the thermal motion of the ions. This gives the liquid charge layer its spatial extension and hence is referred to as a diffuse layer. Figure 2.7 shows the end result which is the formation of a double layer system made up of fixed charges at the surface (Stern layer) and diffuse layer which extends out into the fluid.

![Diagram of a surface with a positive charge with counterbalancing negative ions in the Stern and Diffuse layers.](image)

**Figure 2.7.** Surface with a positive charge with counterbalancing negative ions in the Stern and Diffuse layers.
2.4.2.1 Diffuse Part of Electric Double Layer

The diffuse part of the double layer will be considered first. The simplest quantitative treatment of the diffuse part was developed by Gouy and Chapman. It is based on the following assumptions:

1. The surface is flat, infinite in extent and uniformly charged.
2. The ions in the diffuse part of the double layer are assumed to be point charges distributed according to the Boltzmann distribution.
3. The solvent influences the double layer only through its dielectric constant which is assumed to be the same throughout.
4. Only a single symmetrical electrolyte is present.

Consider when the electric potential at the surface is \( \psi_s \) and \( \psi \) at a distance \( x \) away from the surface in an electrolyte solution. If the surface is positively charged as depicted in Figure 2.8 and by applying the Boltzmann distributions for positive and negative ions we have

\[
    n_+ = n_o \exp \left( -\frac{z^* e \psi}{kT} \right) \quad \text{and} \quad n_- = n_o \exp \left( \frac{z^* e \psi}{kT} \right) \tag{2.28}
\]

where \( n_+ \) and \( n_- \) are the respective numbers of positive and negative ions per unit volume at a given potential \( \psi \), \( n_o \) is the corresponding bulk concentration of the ionic species, \( z^* \) is electrolyte valance charge, \( e \) is the elementary electric charge, \( k \) is the Boltzmann constant and \( T \) is the absolute temperature.
Figure 2.8. Schematic representation of a diffuse electric double layer.

The net volume charge density $\rho$ at points where the potential is $\psi$ is therefore given by

$$\rho = z^* e (n_+ - n_-) = z^* en_o \left( \exp \left( -\frac{z^* e \psi}{kT} \right) - \exp \left( \frac{+z^* e \psi}{kT} \right) \right) = -2z^* en_o \sinh \left( \frac{z^* e \psi}{kT} \right)$$

$\rho$ is related to $\psi$ by Poisson’s equation, which for a flat double layer takes the form

$$\frac{d^2 \psi}{dx^2} = -\frac{\rho}{\varepsilon}$$

(2.30)

where $\varepsilon$ is the permittivity. Combining Eqs. 2.29 and 2.30 yields
\[
\frac{d^2 \psi}{dx^2} = \frac{2z^*e}{\varepsilon} \sinh \left( \frac{z^*e\psi}{kT} \right)
\]

which is known as the Poisson-Boltzmann expression. Solving this equation with the proper boundary conditions will enable one to determine the local electric double layer potential \( \psi \) at a given distance \( x \) away from the surface. If the surface potential is low \( (z^*e\psi_s/kT \ll 1) \) Eq. 2.31 can be solved to give the simple result of

\[
\psi = \psi_s \exp(-\kappa x)
\]

\[
\kappa = \sqrt{\frac{e^2}{\varepsilon_\varepsilon_o kT} \sum n_i^* z_i^*}
\]

where \( \kappa \) is the Debye-Hückel parameter of the electrolyte solution, \( \varepsilon_\varepsilon \) is the dielectric constant of the medium, and \( \varepsilon_\varepsilon_o \) is the dielectric permittivity of a vacuum \( (\varepsilon = \varepsilon_\varepsilon_o) \) and \( n_i^* \) is the electrolyte concentration. Eq. 2.32 shows that for low potentials the potential decreases exponentially with distance from the charged surface. The Debye-Hückel parameter determines the extent the diffuse layer extends into the solution. At a distance of \( 1/\kappa \) from the surface the potential has fallen to a value of \( 1/\exp(1) \) of the surface potential. For this reason, the length \( 1/\kappa \) is known as the Debye length or thickness of the diffuse layer.

**2.4.2.2 Inner Part of Electric Double Layer (Stern Layer)**

The treatment of the diffuse part of the double layer by Gouy (1910) and Chapman (1913) was based on an assumption of point charges in the electrolyte medium. In reality, ions in solution are not point charges but have a finite size. Hydrated ions have diameters typically on the order of 0.5 nm or less. This allowance for the size was an
important correction to Gouy-Chapman theory which was first proposed by Stern. Because ions are hydrated the charge they possess cannot touch the interface of the surface. Instead they are limited to a distance of approach equal to the hydrated radius of the ion. The distance is usually given the symbol $\delta$ as it defines a region which is inaccessible to counterion charge. The plane distance $\delta$ is known as the Stern plane and the regions between this plane and the surface is known as the Stern layer. Outside the Stern layer is a diffuse layer of counterions, just as in the Gouy-Chapman model. Therefore, the combination of the Stern layer and diffuse layer forms the electric double layer which is the basis for the present work. Figure 2.9 shows the entire structure of the electric double layer where the potential drops linearly within the Stern layer because of a constant electric field and then exponentially within the diffuse layer (for low potentials).

Figure 2.9. Schematic representation of the structure of the electric double layer which incorporates Stern’s theory.
2.4.2.3 Zeta Potential

The potential at the Stern plane, \( \psi_d \), is of great interest. However, there is no direct method for determining the Stern potential. This value is therefore estimated from zeta potential measurements which can be readily determined experimentally. As shown in Figure 2.9, the zeta potential, \( \zeta \), is the potential at the surface of shear (hydrodynamic interface) where liquid molecules can shear off from the surface. The surface of shear is a small distance further away from the Stern plane by several layers of liquid molecules and is marginally smaller in magnitude than \( \psi_d \). In double layer theory it is customary to assume identity between \( \psi_d \) and \( \zeta \) and the bulk of experimental evidence suggests that errors introduced through this assumption are usually small, Shaw (1992).

2.4.2.4 Force between Two Electric Double Layers

When two surfaces approach each other the interpenetration of double layers of like sign will lead to repulsion. The force of repulsion can be determined by calculating the potential energy of interaction between two double layers and taking the derivative of the interaction energy, \( E \), with respect to separation distance, \( H \), \( F_{el} = -dE_{el}/dH \). The details of this can be found in the following works [Hogg et al. (1965); Ohshima et al. (1982); Verwey & Overbeek (1999); Butt, Graf and Kappl (2003); Hunter (2001)].

Once the interaction energy between two flat surfaces is determined the interaction energy involving a curved surface can be ascertained. This is done by using Derjaguin’s approximation which assumes the interaction between two double layers on spherical surfaces to be made up of contributions from infinitesimally small parallel rings each of which can be considered as a flat plate. This work was first done by Hogg et al.
(1965) for low surface potentials. Oshima et al. (1982) then extended their work for use at higher surface potentials which can accurately express the interaction energy of approaching double layers.

The analytical expressions developed by Oshima et al. which allow one to calculate the interaction energy, $V_{sp3}$, between two spheres are given below in Figure 2.10

\[ V_{sp1} = \frac{8\pi a_1 a_2 kT}{\kappa^2(a_1 + a_2)} \left[ Y_+^2 \log_e(1 + e^{-\kappa H}) + Y_-^2 \log_e(1 - e^{-\kappa H}) \right], \quad [49] \]

\[ V_{sp2} = V_{sp1} + \frac{8\pi a_1 a_2 kT}{\kappa^2(a_1 + a_2)} \left[ -\frac{1}{48} \left( Y_+^4 + 3Y_+^2 Y_-^2 \right)(\kappa H/2) \{ 1 - \tanh(\kappa H/2) \} \right. \]
\[ \left. + \frac{1}{48} \left( Y_+^4 + 3Y_+^2 Y_-^2 \right)(\kappa H/2) \{ \coth(\kappa H/2) - 1 \} \right], \quad [50] \]

\[ V_{sp3} = V_{sp2} + \frac{8\pi a_1 a_2 kT}{\kappa^2(a_1 + a_2)} \left[ -\frac{Y_+^2}{5760} \left( \frac{15}{8} Y_+^2(7Y_+^2 + Y_-^2) \right)(\kappa H/2) \{ 1 - \tanh(\kappa H/2) \} \right. \]
\[ \left. + \frac{17}{46080} \frac{4(\kappa H/2) \tanh(\kappa H/2)}{\cosh^2(\kappa H/2)} - \frac{Y_+^2}{46080} \frac{4(\kappa H/2) \coth(\kappa H/2) + 17}{\sinh^2(\kappa H/2)} \right] \]
\[ + \frac{Y_+^4 Y_-^2}{1024} \frac{1 + (\kappa H/2) \tanh(\kappa H/2)}{\cosh^2(\kappa H/2)} - \frac{Y_+^4}{1024} \frac{(\kappa H/2) \coth(\kappa H/2) + 1}{\sinh^2(\kappa H/2)} \]
\[ - \frac{Y_+^4}{15360} \frac{1 - 11(\kappa H/2) \tanh(\kappa H/2)}{\cosh^4(\kappa H/2)} + \frac{Y_+^6}{15360} \frac{11(\kappa H/2) \coth(\kappa H/2) - 1}{\sinh^4(\kappa H/2)} \]
\[ + \frac{(\kappa H/2)^2}{1536} \left( \frac{Y_+^2}{\cosh^2(\kappa H/2)} - \frac{Y_-^2}{\sinh^2(\kappa H/2)} \right) \right], \quad [51] \]

Figure 2.10. Analytical expressions by Oshima et al. as taken directly from their work, Oshima et al. (1982). Total electrostatic interaction energy is given as $V_{sp3}$.  

where $a_1$ and $a_2$ are the radii of the two spheres, $n$ is the electrolyte concentration ($n^*$ in the rest of the text), $k$ is the Boltzmann constant, $T$ is the absolute temperature, and $\kappa$ is the Debye-Hückel parameter previously given in Eq. 2.15 and $H$ is the minimum separation distance. The other parameters are defined as

$$Y_+ = \frac{Y_{s1} + Y_{s2}}{2}, \quad Y_- = \frac{Y_{s1} - Y_{s2}}{2}$$

(2.34)

$$y_{s1} = \frac{z^* e}{kT} \psi_{s1}, \quad y_{s2} = \frac{z^* e}{kT} \psi_{s2}$$

where $\psi_{s1}$ and $\psi_{s2}$ are the respective surface potentials involved, $z^*$ is the electrolyte valance charge and $e$ is the elementary electric charge. The force between a sphere and a flat plate caused by two approaching double layers can be obtained by differentiating Eqs. 49-51 from Oshima et al. with respect to $H$ and taking the limit as one of the radii goes to infinity. The results are quite lengthy and are therefore only the first term is provided below

$$F_{el} = \frac{4\pi d \rho n k T e^{-\kappa H}}{\kappa} \left[ \frac{Y_+^2}{1 + e^{-\kappa H}} - \frac{Y_-^2}{1 - e^{-\kappa H}} \right]$$

(2.35)

This expression, however, is only valid for the conditions given in Eq. 2.36 as a result of using Derjaguin’s approximation.

$$H << a$$

(2.36)

$$ka >> 1$$

The force of repulsion between two like double layers is determined by substituting the zeta potentials $\zeta_1$ and $\zeta_2$ for the surface potentials $\psi_{s1}$ and $\psi_{s2}$. 
2.4.3 Potential Energy Curves

In the classical DLVO theory developed by Derjaguin, Landau, Verwey and Overbeek the total force between a particle and wall is the sum of the repulsive electrostatic forces and attractive van der Waals forces. It is common to describe the force balance between \( F_{vdw} \) and \( F_{el} \) in the form of a potential energy-distance diagram shown in Figure 2.11. The total potential energy of interaction, \( E_t \), is given as

\[
E_t(H) = E_{vdw}(H) + E_{el}(H)
\]  

where \( E_{vdw} \) and \( E_{el} \) are the energies associated with the van der Waals and electric double layer interactions, respectively.

![Potential Energy-distance diagram](image)

**Figure 2.11. Potential Energy-distance diagram.**
Equations 2.38A and 2.38B give the interaction energies for the van der Waals and electrostatic forces between a particle and a flat plate. These are the formulas from which Eqs. 2.25 and 2.35 were derived.

\[
E_{vdw}(H) = -\frac{A_{122}d_p}{12H} \left( 1 - \frac{5.32H}{\lambda_c} \ln \left( 1 + \frac{\lambda_c}{5.32H} \right) \right) \quad (2.38A)
\]

\[
E_{el}(H) = \frac{4\pi d_p n kT}{\kappa^2} \left( Y_+^2 \ln(1 + e^{-\lambda_H}) + Y_-^2 \ln(1 - e^{-\lambda_H}) \right) \quad (2.38B)
\]

Additionally, the interaction energies for two particles of diameter \(d_1\) and \(d_2\) which are approaching each other are given by Eqs. 2.39A and 2.39B as

\[
E_{vdw}(H) = -\frac{A_{122}d_1d_2}{12(d_1 + d_2)H} \left( 1 - \frac{5.32H}{\lambda_c} \ln \left( 1 + \frac{\lambda_c}{5.32H} \right) \right) \quad (2.39A)
\]

\[
E_{el}(H) = \frac{4\pi d_1d_2 n kT}{\kappa^2(d_1 + d_2)} \left( Y_+^2 \ln(1 + e^{-\lambda_H}) + Y_-^2 \ln(1 - e^{-\lambda_H}) \right) \quad (2.39B)
\]

As a result of the differences in the potential energy-distance relationships for the van der Waals and electric double layer interactions, the total energy shows a maximum at short separation distances provided the force of repulsion is sufficiently large. In fouling, this maximum represents an energy barrier to the transport of a particle to a heat exchanger wall. Potential energies are often expressed in units of \(kT\). Particles will rarely have energy of more than a few \(kT\). Therefore, a barrier height of 10\(kT\) or more implies a very stable situation where only a minute fraction of the particles will have sufficient energy to surmount the barrier, Van de Ven (1989). If the barrier is overcome, then a particle will be held by van der Waals attraction in the deep primary minimum where removal is less likely.
According to DLVO theory higher surface potentials and a lower ionic strength of the fluid are desirable. This will aid in raising the energy barrier which should in principle help to minimize fouling. Figures 2.12 and 2.13 show the effect of changes in stern potential, $\psi_d$, and Debye-Hückel parameter, $\kappa$, on the potential energy of interaction between two spherical particles. In reality, the mitigation of fouling is more complex because other interactions are possible.

**Figure 2.12.** The influence of Stern potential, $\psi_d$, on the total potential energy of interaction of two spherical particles, Shaw (1992). Particle radii = $10^{-7}$ m, $T= 298$ K, $z^* = 1$, $A_{11} = 2 \times 10^{-19}$ J, $A_{33} = 0.4 \times 10^{-19}$ J, $\varepsilon/\varepsilon_0 = 78.5$, $\kappa = 3 \times 10^8$ m$^{-1}$.  

![Figure 2.12. The influence of Stern potential, $\psi_d$, on the total potential energy of interaction of two spherical particles, Shaw (1992). Particle radii = $10^{-7}$ m, $T= 298$ K, $z^* = 1$, $A_{11} = 2 \times 10^{-19}$ J, $A_{33} = 0.4 \times 10^{-19}$ J, $\varepsilon/\varepsilon_0 = 78.5$, $\kappa = 3 \times 10^8$ m$^{-1}$.](image-url)
Figure 2.13. The influence of electrolyte concentration, $\kappa$, on the total potential energy of interaction of two spherical particles, Shaw (1992). Particle radii $= 10^{-7}$ m, $T= 298$ K, $z^* = 1$, $A_{11} = 2 \times 10^{-19}$ J, $A_{33} = 0.4 \times 10^{-19}$ J, $\varepsilon/\varepsilon_0 = 78.5$, $\psi_d=50$ mV.

2.4.4 Effect of pH on Zeta Potential and Fouling

pH is the variable which is most often used to control the zeta potential. Figure 2.14 shows a typical zeta potential vs. pH curve for silica and alumina. $\zeta$ for nearly all materials becomes negative at higher pH and positive at lower pH values due to the interaction of the potential determining ions in solution ($H^+$ and $OH^-$) that can affect chemical equilibria at the surface. Important in all of this is the fact that interpenetration of double layers of like sign (like zeta potentials) can lead to repulsion which, if large enough, will minimize fouling behavior. The work of Newson et al. (1988) demonstrated this fact. They studied the deposition of haematite particles ($Fe_2O_3$) of $\approx 0.2$ µm in size.
from a water system onto aluminum and 316 stainless steel tubes. Figure 2.15 shows the effect of pH on the measured haematite deposit mass per unit area with time. In their experiments the particulate concentration was 100 mg/kg or 100 ppm with a Reynolds number of 11,000 (turbulent flow).

Figure 2.14. Variation of zeta potential as a function of pH for silica and alumina, Holmber, (2001).

Figure 2.15. Effect of pH on haematite deposition, Newson et al. (1988).
2.5 Hydrodynamic Forces

Hubbe (1984) came up with a detailed analysis of the detachment of particles from a flat surface exposed to a linear shear flow. He showed that the component of hydrodynamic force acting parallel to the wall is usually much larger than the normal force. Consequently, he surmised that drag force is the critical or rate-determining step in the process of re-entrainment of a particle. Sharma et al. (1992) and Yiantsios and Karabelas (1995) later demonstrated that for particles in the size range between 2 to 40 microns, particle removal is controlled by rolling off the surface and not by lift nor some other mechanism such as sliding. The detachment is the result of a torque that is exerted on particle by the viscous drag (see Figures 2.17A and B). Once the restraining torque, determined by the balance between the van der Waals and electrostatic forces is overcome the particle will start to roll and will be entrained in the flow.

With the above discussion in mind, there are two hydrodynamic forces that can have a major impact on fouling behavior. The first is drag force which is responsible for particle removal and the second is lift force which can prevent a particle from adhering to a surface in the first place. Both of these are related to the shear stress at the wall which can be observed in Eqs. 2.40 and 2.45. Hence, it is the wall shear stress which is often regarded as the true removal force.

2.5.1 Hydrodynamic Lift Force between Particle/Wall

When a particle is in near contact with a fixed plane wall as fluid flows by, a lift force on the particle is created. This is caused by the fact that the fluid further away from the wall is at a higher velocity and hence at a lower pressure. It is this pressure difference
that results in the lift force. This situation is depicted in Figure 2.16. In a practical system this lift force may prevent a particle from making intimate contact with a heat exchanger surface and thereby mitigate fouling.

![Figure 2.16. Particle in contact with planar surface while in a linear shear flow.](image)

The hydrodynamic lift force for a spherical particle in the vicinity of the deposition surface is given by Leighton and Acrivos (1985) who extended the analysis of O’Neill (1968) by taking into account weak inertial effects. They found that the lift force, $F_L$, on a spherical particle in contact with a horizontal wall, in a slow linear shear flow is

$$ F_L = 0.58 \rho_f \left( \frac{\tau_w}{\mu_f} \right)^2 d_p^4 $$  \hspace{1cm} (2.40)

This expression is valid for sufficiently small particle Reynolds numbers ($\text{Re}_p < 1$) such that the inertial terms in the Navier-Stokes equations can be neglected. In addition, it can be used to closely approximate the lift force with a separation distance of tens of nanometers. The equation for calculating the particle Reynolds number is given below

$$ \text{Re}_p = \frac{2R_p V_{y=R}}{\nu} $$  \hspace{1cm} (2.41)
where $R_p$ is the radius of the particle and $V_y=R_p$ is the velocity of the fluid at a distance $R_p$ from the wall. Under laminar flow conditions the velocity profile is parabolic and not linear as depicted in Figure 2.16. However, at very small distances away from the wall the profile may be considered linear. The wall shear stress is calculated as

$$\tau_w = \frac{1}{2} f \rho f V_m^2$$  

where $f$ is the Fanning friction factor. The friction factor is calculated from the Poiseuille number which is a function of the aspect ratio for a rectangular channel. It can be determined using Eq. 2.42 from Shah and London (1978).

$$f \text{ Re} = 24 \left(1 - 1.3553 \alpha_c^* + 1.9467 \alpha_c^{*2} - 1.7012 \alpha_c^{*3} + 0.9564 \alpha_c^{*4} - 0.2537 \alpha_c^{*5}\right)$$  

where $\alpha_c^*$ is the channel aspect ratio. One constraint for this equation is that the aspect ratio must be less than one. If the channel aspect ratio is greater than unity, the inverse is taken for use in Eq. 2.43.

### 2.5.2 Hydrodynamic Drag Force on Particle Attached to Wall

The drag force on a spherical particle of radius $R_p$ in contact with a plane wall in a slow linear shear flow was calculated by O’Neill (1968). His results yield

$$F_D = 1.7(6\pi) \mu_f R_p V_{y=R_p}$$  

(2.44)

This force may be expressed in terms of the wall shear stress.

$$F_D = 32R_p^2 \tau_w = 8d^2 \tau_w$$  

(2.45)

If the release mechanism is rolling, then the torque balance is given as

$$F_A \cdot r_a = 1.399 F_D \cdot R_p$$  

(2.46A)
where $F_A$ is the total adhesive force from DLVO forces and $r_a$ is the radius of contact area between the particle and the wall as shown in Figure 2.17A for a deformable particle. The factor 1.399 comes from Sharma et al. (1992) who determined that the hydrodynamic force acting over the surface can be replaced by an effective force acting at a distance $1.4R_p$ from the wall. When the drag force is great enough, the restraining torque will be overcome and the particle will begin to roll and subsequently be entrained in the flow. When the particle is non-deformable the contact radius may be replaced with a lever arm, $L$. This situation is depicted in Figure 2.17B where there is a characteristic height of roughness, $h$. It can be shown from the figure that

$$L = R_p \sin \beta = R \sqrt{\frac{2h}{R_p} + \frac{h^2}{R_p^2}}$$

(2.46B)

It is evident that the surface roughness affects the ability of a particle to become detached from a surface. In short, a greater surface roughness requires more shear stress at the wall in order for a particle to detach.

Figure 2.17A. Schematic diagram of a torque balance for a deformable particle with a contact radius of $r_a$. Deformation of particle is greatly exaggerated.
2.6 Force of Gravity

If a particle is large enough the force of gravity on a particle may be significant enough to influence fouling behavior. The gravitation force, is however negligible for nano-sized particles in comparison to the electrostatic and van der Waals forces that are typically present. However, if particles aggregate to tens of microns it can be an important factor. The force of gravity, \( F_g \), on a particle immersed in a fluid is given as

\[
F_g = \frac{\pi d_p^3 (\rho_p - \rho_f) g}{6}
\]  

(2.47)

where \( g \) is the acceleration due to gravity.
2.7 Pressure Drops in Microchannels

The microchannels under consideration are to be used as a heat exchanger for dissipating thermal energy. For this reason, they will function with greater efficiency if placed in a parallel array and connected together with an inlet and exit manifold as shown in Figure 2.18. Therefore, the present work focuses on single-phase liquid flow carrying solid particulates which may lead to fouling and give rise to an increased pressure drop across the channels.

![Image of silicon, parallel microchannels with inlet and outlet manifolds. Microchannels are 10 mm in length and together have a width of 8 mm.](image)

**Figure 2.18.** Photograph of silicon, parallel microchannels with inlet and outlet manifolds. Microchannels are 10 mm in length and together have a width of 8 mm.

Compressibility effects and slip boundary conditions do not apply for the single phase liquid flow used in this work. In addition, the continuum assumption is known to be valid for liquid flows within microchannels for hydraulic diameters greater than 1 µm (which is the case here), Gad-el-Hak (1999). With the above discussion in mind, the basic
theory of pressure drops within microchannels will be covered. This is necessary since the fouling phenomena being studied are related to the increase in pressure drop which can occur within a microchannel device.

2.7.1 Friction Factor Theory

A one-dimensional flow in a smooth circular pipe will form the starting point for this analysis. The following equations are readily derived based on the continuum assumption for Newtonian liquids. A control volume of length dx is shown in Figure 2.19 where D is the pipe diameter.

The pressure over the inlet area is \( p - \frac{dp}{2} \), and over the outflow area is \( p + \frac{dp}{2} \). Since the flow being considered is fully developed, it is in equilibrium under the applied forces (i.e., no change in momentum, fluid is not being accelerated). The pressure acts over the cross-sectional area \( \pi D^2/4 \), and the viscous stress acts on the surface area \( 2\pi Ddx \). A force balance on the fluid element gives

\[
\left( p - \frac{1}{2} dp \right) \frac{\pi D^2}{4} - \left( p + \frac{1}{2} dp \right) \frac{\pi D^2}{4} = \tau_w \pi D dx
\]

so that

\[
(2.48)
\]
\[
\frac{dp}{dx} = -\frac{4}{D} \tau_w
\]  \hspace{1cm} (2.49)

Equation 2.49 shows that the pressure gradient (pressure drop over distance) is related to the wall shear stress. Given a Newtonian fluid the wall shear stress, \( \tau_w \), is expressed in terms of the velocity gradient at the wall as

\[
\tau_w = \mu f \left( \frac{du}{dy} \right)_w
\]  \hspace{1cm} (2.50)

where \( u \) is the velocity at a given distance \( y \) normal to the wall. The Fanning friction factor \( f \) is defined as

\[
f = \frac{\tau_w}{\frac{1}{2} \rho f V_m^2}
\]  \hspace{1cm} (2.51)

The frictional pressure drop \( \Delta P \) over a length \( L \) is obtained from Eqs. 2.49 and 2.51 giving

\[
\Delta P = \frac{2 f \rho f V_m^2 L}{D}
\]  \hspace{1cm} (2.52)

For non-circular flow channels \( D \) in Eq. 2.52 is replaced with the hydraulic diameter \( D_h \) which is defined by the following equation

\[
D_h = \frac{4 A_c}{P_w}
\]  \hspace{1cm} (2.53)

where \( A_c \) is the channel cross-sectional area and \( P_w \) is the wetted perimeter. Using this definition, the hydraulic diameter of a circular pipe will be equal to \( D \), the inner pipe diameter. For a rectangular channel of sides \( a \) and \( b \), \( D_h \) is given by

\[
D_h = \frac{2ab}{(a + b)}
\]  \hspace{1cm} (2.54)
In laminar flow, the Poiseuille number, \( \text{Po} = \frac{f}{\text{Re}} \) is a constant. The \( f\text{Re} \) value for a circular pipe is 16. However, the Poiseuille number for a rectangular passage is a function of its aspect ratio. This was given earlier in Eq. 2.43 and is restated here

\[
f \text{Re} = 24\left(1 - 1.3553\alpha_c^* + 1.9467\alpha_c^{*2} - 1.7012\alpha_c^{*3} + 0.9564\alpha_c^{*4} - 0.2537\alpha_c^{*5}\right)
\]

where \( \alpha_c^* \) is the channel aspect ratio. The above discussion assumes laminar flow where the fluid particles move in smooth layers or lamina. This is the case when the Reynolds number is less than 2100 for smooth circular tubes. Reynolds number is defined as

\[
\text{Re} = \frac{\rho f V_m D_h}{\mu_f}
\]

which is the ratio of inertial forces to viscous forces. Equation 2.52 can be rearranged to be a function of the Poiseuille number, \( f\text{Re} \), which is more useful for engineering calculations. It has the following form

\[
\Delta P = \frac{2(f \text{Re})\mu_f V_m L}{D_h^2}
\]

### 2.7.2 Developing Laminar Flow and Boundary Layer

The hydrodynamic developing flow can be very important in microchannels. Due to the often short lengths employed, it is possible for the developing flow to dominate over the entire length of the microchannel. Therefore, it is considered here.

Whenever a fluid is forced to flow along a solid surface (boundary), a boundary layer is formed as a result of the fluid velocity being zero at the surface. This has been determined experimentally and is referred to as a no-slip condition. In fluid mechanics, the boundary layer is defined as the portion of the fluid whose velocity is appreciably affected by the presence of a solid surface. It is often defined as the region where the
fluid velocity is less than 99% of the free stream velocity. It is the formation of this boundary layer which gives rise to the developing flow in a channel.

Figure 2.20 illustrates laminar flow in the entrance region of a conduit. The flow starts out uniform at the inlet and because of the no-slip condition at the wall the velocity profile changes. Eventually, downstream the boundary layers merge. A short distance later the velocity profile no longer changes. At this point the flow is referred to as being fully developed and has a parabolic profile when observed in a single dimension.

![Figure 2.20. Laminar flow in the entrance of a channel, Crowe (2001).](image)

The length of the hydrodynamic developing region, $L_e$, is given by the following well established, empirical equation

$$L_e = 0.05 \text{Re} D_n$$

(2.57)

Therefore, if $L_e$ is less than the channel length the pressure drop within the flow passage can be determined with help from Eq. 2.56.

To account for the developing region of a channel, the pressure drop equations are often presented with an apparent friction factor, $f_{\text{app}}$, in place of the Fanning friction factor. The apparent friction factor accounts for both the pressure drop due to friction
and the developing flow at the entrance of the channel. Using this averaged value, the pressure drop equation can be rewritten as

$$\Delta P = \frac{2(f_{\text{app}} \text{Re}) \mu_j V_m x}{D_h^2}$$  \hspace{1cm} (2.58)

where \(x\) is the length down the channel. The difference between the apparent friction factor over a length \(x\) and the fully developed friction factor is expressed in terms of an incremental pressure defect \(K(x)\)

$$K(x) = \left( f_{\text{app}} - f \right) \frac{4x}{D_h}$$  \hspace{1cm} (2.59)

When \(x > L_e\) the incremental pressure defect obtains a constant value \(K(\infty)\), known as Hagenback’s factor. The total pressure drop which accounts for the developing flow is obtained by combining Eq. 2.58 and 2.59 to give

$$\Delta P = \frac{2(f \text{Re}) \mu_j V_m x}{D_h^2} + K(x) \frac{\rho_j V_m^2}{2}$$  \hspace{1cm} (2.60)

Like the Poiseuille number the Hagenback factor has a dependence on the channel aspect ratio for rectangular channels. It is given as

$$K(\infty) = 0.6796 + 1.2197 \alpha_e^* + 3.3089 \alpha_e^{*2} - 9.5921 \alpha_e^{*3} + 8.9089 \alpha_e^{*4} - 2.9959 \alpha_e^{*5}$$  \hspace{1cm} (2.61)

This equation was developed by Steinke and Kandlikar (2005) and has an accuracy of 0.04%.

2.7.3 Total Pressure Drop in a Microchannel Heat Exchanger

There are several components that contribute to the total pressure drop within a microchannel. These are (1) the fully developed frictional losses, (2) the losses from the hydrodynamic developing length and, (3) the inlet and exit losses. The different
The components of the pressure loss are illustrated in Figure 2.21 where the inlet and outlet losses are represented by their coefficients $K_i$ and $K_o$, respectively.

![Figure 2.21. Components of pressure drop when measuring $ΔP$ in a microchannel which has inlet and outlet plenums/manifolds, Steinke (2005).](image)

The total pressure drop in this case is

$$ΔP = \frac{2(f \, Re) \mu_{nf} V_m x}{D_n^2} + K(x) \frac{ρ_f V_m^2}{2} + K_o \frac{ρ_f V_m^2}{2}$$

(2.62)

The first term represents the losses from fully developed flow. The second term comes from the frictional losses due to the hydrodynamic developing region and the last two terms correspond to the losses taking place at the inlet and exit of a channel. The inlet and exit losses are often referred to as minor losses since their contribution to the total is
small. These losses are attributed to flow separation where the fluid does not flow smoothly in the intended direction. Significant parts of the fluid can eddy and recirculate in a manner that absorbs mechanical energy without doing useful work.

2.8 Particle Interception

Particle interception can be an important concept in particulate fouling. This is especially true when dealing with a heat exchanger with parallel microchannels connected with an inlet manifold. In this phenomenon, a particle can collide with the cross-sectional area of a fin structure and be collected (become trapped), after which additional particles can be amassed until significant blockage has occurred at the channel entrances. After significant build up, the fluid flow into the channels can be impeded by a reduction in the cross-sectional area at the channel inlets. In addition, it is possible for this phenomenon to have a larger impact when the fins make up a larger contribution of the total cross-sectional area. Figure 2.22 helps illustrate this point.

![Figure 2.22. Cross-sectional view of parallel channels. The top channels (A) have a greater fin cross section than the bottom channels (B) and hence may be more susceptible to fouling by particle interception at the channel inlets.](image.png)
2.9 Maldistribution and Channel Constriction

The pressure drop in a multichannel network can be influenced by the maldistribution of fluid into the channels. This unequal distribution of fluid can be caused by the blockage of some of the channels with foulant. The end result is increased flow velocities in the other channels and hence a greater pressure drop.

If the channels are constricted along their length or simply at the inlets, significant pressure drops can arise. For a circular pipe, the pressure drop can be expressed as

$$\Delta P = \frac{128 \mu_L L Q}{\pi D^4}$$

where $Q$ is the volumetric flowrate. The above equation shows that for a constant flowrate, a constriction of the diameter by 5% will result in a pressure drop increase of 23%. This same heavy dependency on the pipe diameter also applies to the inlet losses which have been rewritten below.

$$\Delta P_{\text{inlet}} = \frac{8K_i \rho_i Q^2}{\pi^2 D^4}$$

2.10 Investigations in Fouling/Particle Deposition

There are some general aspects in fouling or particle deposition studies which are common to many experimentalists working in this area. This section helps to cover (1) how fouling is quantified, (2) what types of general instrumentation or analyses are performed, and (3) how randomness in fouling is addressed.
2.10.1 Quantification of Fouling/Particle Deposition

As was discussed at the beginning of this chapter, fouling has almost exclusively been quantified by the increase in thermal resistance to heat transfer. However, some researchers have studied fouling/particle deposition in terms of the particle deposition coefficients, particle deposition/removal rates and the fraction of particles removed while under quantifiable hydrodynamic and physiochemical forces. Other researchers have measured fouling in terms of the mass of the foulant per unit area.

It is however rare for the pressure drop to be used as a figure of merit in fouling studies. If it is ever addressed, it is only briefly mentioned in a paper. The one exception to this is the work done by Masri and Cliffe (1996). Their experiments explored the deposition of fine particles in compact fin heat exchangers. The hydraulic diameters used in their work were relatively small and hence were susceptible to increases in pressure drop. Figure 2.23 illustrates the pressure drops they observed for wavy fin minichannels with a hydraulic diameter of 2.39 mm.
Figure 2.23. Normalized pressure drop for a wavy fin geometry used in a compact fin heat exchanger. Redrawn from work of Masri and Cliffe (1996).

2.10.2 General Instrumentation/Analyses

The use of a microscope is by far the most common tool used in fouling/particle deposition studies. It provides good visualization of the growth of the foulant layer. Yiantsios and Karabelas (2003) used a dark-field illumination microscope with its excellent contrasting capability. Figure 2.24 shows a microscopy image of their work. Here silica particles on a glass substrate are sharply contrasted thus allowing for the particles to be numbered for the calculation of particle deposition coefficients.
Another common tool used is a Scanning Electron Microscope (SEM). This has been used to observe the crystal structure formed during crystallization fouling. In addition, X-ray diffraction has been used to determine the elements that are present in some unknown foulant materials within crude oil as well as dusts in air heat exchangers.

2.10.3 Addressing Randomness in Fouling

A certain degree of scatter or randomness is commonly accepted in fouling phenomena. This may be attributable to the partial removal of deposits over time followed by a rapid build up of material or the rearrangement or non-uniformity of deposition of the fouling material. Figures 2.25 and 2.26 both help illustrate this aspect of fouling. Figure 2.25 demonstrates the type of scatter that can be expected. Figure 2.26 shows that for different operating conditions, the fouling observed can at times overlap each other or have similar values. In this figure the fouling resistance at 200 and 260°C intersect each other after 60 hours of operation. In short the data can be somewhat “messy” to analyze.
Figure 2.25. Graph demonstrates the random behavior that can exist in fouling studies. The run for no filtration exhibits this fact the best, Bansal et al. (2001). Original data was to demonstrate the effect of filtration (1 µm) on calcium sulfate fouling (crystallization fouling).

Figure 2.26. Graph shows that for different operating conditions (heat exchanger surface temperature, $T_{s,o}$) the results can be very similar such that little differentiation can be made in the data, Saleh et al. (2005). Original data was to demonstrate the effect of initial surface temperature on fouling resistance.
The use of statistics has also been used to develop definitive answers in fouling studies. In addition to using the mean and standard deviation of the data, statistical t and F tests have been employed. In both tests, it is assumed that the samples are random and come from normally distributed populations. These statistical tests are well established and, as such, detailed explanations of them can be found in any standard statistics book.

A t-test assesses whether the means of two groups are statistically different from one another. It utilizes the mean, $\mu$, standard deviation, $\sigma$, variance, $\sigma^2$, and sample number, $n$. The test statistic is given as

$$t_o = \frac{\mu_1 - \mu_2}{\sigma_p \sqrt{\frac{1}{n_1} + \frac{1}{n_2}}}$$

where the subscripts denote group 1 and 2, respectively. An estimate of the common variance among the two samples, $\sigma_p^2$, is given as

$$\sigma_p^2 = \frac{(n_1 - 1)\sigma_1^2 + (n_2 - 1)\sigma_2^2}{n_1 + n_2 - 2}$$

The value of $t_o$ is utilized in conjunction with statistical tables to determine if the difference in the means is statistically significant given a certain confidence level. A typical confidence level is between 90 and 95% but smaller values can be utilized if a process is known to have a fair amount of variability and strong conclusions are difficult to make.

Similarly an F-test determines whether the variances of two groups are statistically different from each other. The test statistic is calculated as

$$F_o = \frac{\sigma_1^2}{\sigma_2^2}$$
Like the t-test the value of $F_o$ is utilized in conjunction with statistical tables to determine if the difference in the variances is statistically significant given an appropriate confidence level.

2.11 Effect of Fibrous Material in Particulate Fouling

When studying particulate fouling in microchannels there are confounding effects which can be introduced by fibrous material. The fiber can come from numerous sources including:

1. Buffers used to control pH
2. Contaminated labware
3. The lab environment, where airborne material makes its way into a dispersion
4. Experimenter, natural shedding of skin and hair
5. The experimental system itself

Fibrous material stemming from the experimental setup should not be present. Careful evaluation of the test loop used to study fouling needs to be performed prior to introducing particulates. Deionized water which has been thoroughly filtered with a submicron filtration apparatus should be run through the system extensively to ensure that the test loop does not introduce variability into the system by shedding fibrous material.

Buffers for pH control are another source of fibrous material that has been demonstrated. Anything that becomes apart of the continuous phase of a solution should be filtered prior to formulating a dispersion for test. The presence of fibrous material in microchannel particulate fouling is a viable confounding influence. Its presence therefore within a microchannel device will be addressed.
The interaction between a particle and a fiber in a microchannel device can be best represented by aerosol filtration theory where fibrous filters consisting of a mat of individual fibers are used to filter out airborne impurities. An electron micrograph of a typical glass fiber filter is shown in Figure 2.27. Typical fibrous filers have a relatively high porosity which ranges from 0.6 to 0.999 (between 60 and 99.9% of the filter by volume is absent of material). The fiber sizes range from less than 1 µm to several hundred microns. The materials used for fibrous filters include cellulose, glass, quartz and plastic fibers.

Figure 2.27. Electron micrograph showing a typical microstructure of a fibrous filter, Baron and Klaus (2001).

In the forthcoming sections the mechanisms involved in aerosol capture by single fibers will be discussed and applied to the analogous phenomenon of a particle interacting with a fiber within a microchannel device. This mixed fouling occurrence can lead to increased pressure drops in particulate fouling because a fiber within a microchannel device is capable of collecting particulates. This in turn creates a larger and denser obstruction to flow.
2.11.1 Single-Fiber Efficiency

The study of filtration involves applying classical fluid mechanics to the flow around obstacles. The starting point in characterizing fiber filtration or in the similarly mechanistic phenomenon of particulate fouling in the presence of fibers is the capture of particles by a single fiber. In this basic model the fiber is treated as a cylinder orientated perpendicular to the flow as depicted in Figure 2.28. The single-fiber efficiency is defined as a dimensionless particle deposition rate on a unit length of fiber. As shown in Figure 2.28 the single-fiber efficiency is the fraction of particles approaching the fiber in the region defined by the projected area of that fiber which are ultimately collected on the fiber. Stated another way, the single-fiber efficiency, $\eta$, is defined as the ratio of the number of particles striking a fiber to the number that would strike if the streamlines were not diverted around the fiber. This is shown in Figure 2.29 where a fiber radius $R_f$ removes all the particles contained in a layer of thickness $Y$ from the centerline of the fiber. The single-fiber efficiency is therefore defined as

$$\eta = \frac{Y}{R_f}$$

(2.68)

This definition of single-fiber efficiency assumes that a particle adheres to a fiber on contact (i.e., there is no particle bounce or previously deposited particles on the fiber). This is an obvious oversimplification when attempting to apply aerosol theory to particle/fiber interactions for the present fouling studies. However, it is still a useful tool in understanding the underlying principles involved.
2.11.2 Deposition Mechanisms

There are four basic mechanical mechanisms by which a particle can come into contact with a fiber. They are:

1. Interception
2. Inertial impaction
3. Diffusion
4. Gravitational settling

Each of these four deposition mechanisms is described below, along with the equations that predict the single-fiber efficiency due to that particular mechanism. The single-fiber efficiency, $\eta$, is assumed in a first order approximation to be equal to the sum of the
individual efficiencies from interception, $\eta_{\text{int}}$, inertial impaction, $\eta_{\text{imp}}$, diffusion, $\eta_{\text{diff}}$, and gravitational settling, $\eta_{\text{grav}}$. This is easily expressed as

$$\eta_{\text{total}} = \eta_{\text{int}} + \eta_{\text{imp}} + \eta_{\text{diff}} + \eta_{\text{grav}}$$  \hspace{1cm} (2.69)

2.11.3 Isolated Cylinder Model

In filtration theory, the fibers are treated as an array of non-interacting cylinders. One of the simplest and most effective models is one developed by Kuwabara which is based on the cell model of randomly spaced cylinders. As shown in Figure 2.30 each fiber is surrounded by an independent body of fluid. The polygonal cells are difficult to treat mathematically and the analysis is simplified to cells which are identical concentric cylinders with dimensions shown in Figure 2.31. The packing density, $\alpha$, of a cell is equal to the fiber volume divided by the total volume of the cell in the two dimensional plane.

$$\alpha = \left( \frac{\text{fiber radius}}{\text{cell radius}} \right)^2 = \left( \frac{R_f}{R_c} \right)^2$$  \hspace{1cm} (2.70)

Figure 2.30. Schematic representation of parallel cylinders with the same radius. Cylinders are randomly placed and homogenously distributed, Tolliver (1988).


2.11.4 Kuwabara Flow Field

Kuwabara solved the viscous flow equations of fluid mechanics in two dimensions to give a distribution of velocity around a cylinder transverse to the flow. The boundary conditions used by Kuwabara are zero velocity on the surface of the fiber and zero vorticity (laplacian of the steam function is zero, $\Delta^2 \psi = 0$) on the surface of cell boundary. Kuwabara’s solution to the problem is a two dimensional stream function, $\psi$, which is a function of position only. At any point it is a measure of the air flowing between one point and some arbitrary origin. The Kuwabara flow field is illustrated in Figure 2.32. In the diagram there is no airflow across lines joining points at which $\psi$ is constant; such lines are streamlines. Closely spaced streamlines indicate rapid flow, whereas widely spaced streamlines indicate slow flow. Moreover, $\psi$, can simply be related to the velocity of the fluid by means of the following equations, in Cartesian coordinates,
Figure 2.32. Streamlines calculated according to the cell model of Kuwabara, Brown (1993).

\[ U_x = \frac{\partial \psi}{\partial y}; \quad U_y = -\frac{\partial \psi}{\partial x} \]  \hspace{1cm} (2.71)

and in polar coordinates

\[ U_r = \frac{1}{r} \frac{\partial \psi}{\partial \theta}; \quad U_\theta = -\frac{\partial \psi}{\partial r} \]  \hspace{1cm} (2.72)

Kuwabara’s solution for the stream function is given as

\[ \psi = \frac{U_\infty r \sin \theta}{2Ku} \left( 2 \ln \frac{r}{R_f} - 1 + \alpha + \frac{R_f^2}{r^2} \left( 1 - \frac{\alpha}{2} \right) - \frac{\alpha}{2} R_f^2 \right) \]  \hspace{1cm} (2.73)

where \( U_\infty \) is the fluid velocity within the filter (\( U_\infty = U_\alpha/(1-\alpha) \)), \( U_\alpha \) is the filter face velocity, \( r \) is the radial coordinate, \( \theta \) is the angular coordinate and \( Ku \) is the hydrodynamic factor given as

\[ Ku = -\frac{1}{2} \ln \alpha - \frac{3}{4} + \alpha - \frac{\alpha^2}{4} \]  \hspace{1cm} (2.74)

In the Kuwabara flow model the flow field is independent of the viscosity of the fluid. However, it does scale with the packing density.
2.11.5 Interception

Particle collection by interception is shown in Figure 2.33. Collision by interception occurs when a particle follows the fluid stream lines and happens to come within one particle radius of the fiber surface. When the particle comes in contact with the fiber it is considered to have been captured because of its finite size. The only real deposition or collection mechanism is interception. The other mechanisms are only deviations from this phenomenon. In pure interception, however, it is assumed that a particle has size but no mass and that it follows the streamlines perfectly. In addition, it experiences negligible effects from inertia, gravity and Brownian motion.

![Figure 2.33. Single-fiber collection by interception, Hinds (1999).](image)

In Figure 29, if \( Y \) is taken as the ordinate at which a streamline cuts the cell boundary, \( R_c \), the efficiency due to interception is

\[
\eta_{\text{int}} = \frac{Y}{R_f}
\]

Moreover, when Kuwabara’s stream function is solved for any point on the outer cylinder of the cell model, at \( R_c \), Eq. 2.73 simplifies to
\[ \psi = U_\nu R_c \sin \theta = U_\nu Y \]  \hfill (2.76)

Hence,

\[ \eta_{int} = \frac{\psi}{U_\nu R_f} \]  \hfill (2.77)

The value of \( \psi \) is determined by choosing a streamline which approaches within a distance, \( R_p \), the particle radius of the fiber when \( \theta = \pi/2 \). Hence substituting \( r = R_f + R_p \) in Eq. 2.73 one can solve for the single-fiber efficiency by interception. A simplified form of the following equation is given by Lee and Ramamurthi (1993) as

\[ \eta_{int} = \frac{(1 - \alpha)N_R^2}{Ku(1 + N_R)} \]  \hfill (2.78)

where \( N_R \) is a dimensionless parameter expressed as

\[ N_R = \frac{R_p}{R_f} \]  \hfill (2.79)

The other collection mechanisms are much more complicated in how their single-fiber efficiencies are derived. However, they follow the same basic outline as given above with the exception that they take into account deviations in the stream function that occur due to inertia, gravity and Brownian motion of a particle. These deviations affect the particle’s trajectory and hence the single-fiber collection efficiency.

### 2.11.6 Inertial Impact

Inertial impact of a particle on a fiber occurs when the particle has sufficient inertia that it is unable to adjust quickly enough to abrupt changes in streamlines close to the fiber and therefore collides with the fiber surface. This is illustrated in Figure 2.34.
this phenomenon collision does not occur until the center of the particle reaches the fiber. Consequently, it is not physically true and is to some extent a theoretical construct.

Figure 2.34. Single-fiber collection by inertial impaction, Hinds (1999).

In solving for the particle motion with significant inertia there are no analytical solutions because of the complexity of the flow field. Therefore, the problem is solved numerically. The governing parameter for this mechanism is the Stokes number. It is defined as the ratio of the particle stopping distance against fluid drag to fiber diameter, $D_f$. The Stokes number is given as

$$
Stk = \frac{t_r U_o}{D_f} = \frac{\rho_p d_p^2 U_o}{18 \mu_f D_f}
$$

Single-fiber efficiency via impaction increases with increasing Stokes number because of (1) the greater particle inertia ($d_p$ or $\rho_p$), (2) larger particle velocity and (3) more abrupt curvature of the streamlines caused by a smaller fiber. The single-fiber efficiency is given by Stechkina et al. (1969) as

$$
\eta_{imp} = \frac{(Stk)J}{2Ku^2}
$$

where $J$ is given as
\[ J = (29.6 - 28\alpha^{0.62})N_R^2 - 27.5N_R^{2.8} \]  

(2.82)

Eq. 2.82 is valid under the following conditions: \(0.01 < N_R < 0.4\) and \(0.0035 < \alpha < 0.111\).

Large particles will also have a significant collection by interception as well. However, the sum of \(\eta_{\text{int}}\) and \(\eta_{\text{imp}}\) cannot exceed a theoretical maximum of \(1 + N_R\).

2.11.7 Diffusion

Diffusion is significant only for small particles (in the nanometer regime). Very small particles undergo significant Brownian motion and as such do not follow the streamlines exactly. Instead they continuously diffuse away from them in a direction normal to the concentration gradient. The fluid streamlines for these size particles only represent the average path taken. Figure 2.35 shows how the trajectory of a particle can be altered by significant Brownian motion. Capture can occur even when the distance between a streamline and a fiber are greater than the radius of the particle.

![Figure 2.35. Single-fiber collection by diffusion, Hinds (1999).](image)

Single-fiber efficiency due to diffusion is a function of the dimensionless Peclet number, \(Pe\), which is given as
where $D_B$ is the diffusivity given in Eq. 2.20. It represents the relative magnitudes of the diffusional and convective motions of a particle in the vicinity of a fiber. This mechanism is inversely proportional to the Peclet number and consequently the only mechanism that becomes more prominent for smaller particle sizes. The single-fiber efficiency is given by Lee and Liu (1982) as

$$
\eta_{diff} = 2.58 \left( \frac{1 - \alpha}{Ku} \right)^{1/3} Pe^{-2/3}
$$

(2.84)

### 2.11.8 Gravitational Settling

Particles will settle when traveling at a low velocity while in a gravitational force field. If the settling is large, the particles will deviate from their streamlines and may colloid with the fiber surface. The dimensionless value that is involved in gravitational settling is given as

$$
G = \frac{V_G}{U_\infty} = \frac{d_p^2 \rho_j g}{18 \mu_f U_\infty}
$$

(2.85)

This is simply the ratio of the settling velocity, $V_G$, to the fluid velocity, $U_\infty$. The single-fiber efficiency has been approximated by Davies (1973) as

$$
\eta_{grav} = \frac{G}{1 + G}
$$

(2.86)
CHAPTER 3

EXPERIMENTAL METHODOLOGY

The experiments included in this project focused on particulate fouling of SiO$_2$ and Al$_2$O$_3$ particles within silicon, rectangular microchannels. In this chapter, the components of the experimental flow loop will be discussed in detail which includes the data acquisition (DAQ) equipment used to collect pressure drop data. The materials and equipment used for the experimental runs will also be covered which includes the particle size and zeta potential analyses as well as the fluid dynamic visualization studies using Micro-PIV (Particle Image Velocimetry).

Dispersions that were tested had a significant particle concentration by weight. The weight percent of the particles was much higher than would actually be seen in any microchannel device used for IC chip cooling. Therefore, in a practical way all the tests considered were accelerated. This is why data was able to be collected within only a few hours of operation during the experimental runs.

3.1 Experimental Flow Loop

Particulate fouling was studied using the flow loop schematic given in Figure 3.1. A peristaltic pump (Cole-Parmer Masterflex L/S) was used to drive the fluid which has a rated speed control of ± 0.25%. With a standard Cole-Parmer L/S 14 pump head it can deliver a volumetric flowrate between 1.3 and 130 ml/min. The pump head used a Cole-Parmer L/S 14 tubing with a 1/16-inch diameter made from a spallation free tubing which goes by the trade name STA-PURE. The STA-PURE tubing can operate at an upper continuous pressure of 414 kPa (60 psi).
During initial experimentation, a different type of tubing made of Norprene was used. However, over time it was observed that the Norprene tubing used in the pump was shedding fibrous material into the microchannel inlets which increased the variability in the pressure drop data. Consequently, new peristaltic tubing (STA-PURE type) was used that was spallation free (small particles are not generated from it) to avoid this confounding by the test setup.

Moreover, the peristaltic pump was selected because it is capable of pumping fluids with a high solids content if desired, and can be run dry to flush the system between runs. An advantage of peristaltic pumps is that they do not have gears that can be worn down with time by particulates which can act as an abrasive. An image of the pump head and experimental flow loop is given in Figures 3.2 and 3.3A-C, respectively.

The flow loop with a test device in place had a volume of 7.4 ml which was taken into account when running experiments. The setup used 1/16-inch inside diameter Chemfluor tubing which is made of a perfluoroalkoxy copolymer with excellent physical properties similar to Teflon®, polytetrafluoroethylene (PTFE).

![Figure 3.1. Schematic of experimental flow loop used in microchannel fouling studies. ΔP is a differential pressure transducer.](image-url)
Figure 3.2. Peristaltic pump head with three internal rollers. Circular arrow shows direction of motion to move fluid through experimental system.

The pressure was measured using either a 6.9 kPa (1 psi), 103 kPa (15 psi) or 207 kPa (30 psi) differential pressure transducer. The pressure transducers are a PX-26 series type from Omega. The transducers have a silicon diaphragm that uses a Wheatstone bridge resistor network to measure the deflection. They all have an excitation voltage of 10 V. The 15 and 30 psi pressure sensors have an output voltage from 0 to 100 mV while the 1 psi sensor has an output voltage ranging from 0 to 16.7 mV. All the pressure sensors had a linearity of ± 0.25% FS BFSL (Full Scale Best Fit Straight Line) and a hysteresis and repeatability of 0.2% FS.
Figure 3.3A. Experimental flow loop. DAQ is the data acquisition system.
Figure 3.3B. Close up of experimental test section resting on microscope stand.
3.2 Pump Calibration

The peristaltic pump was calibrated by weighing the amount of fluid collected over a given period of time for certain volumetric flowrates. This was done three times for each flowrate set point where the average and standard deviation were calculated. The calibration curve is presented in Figure 3.4 which shows a linear correlation.
Since a peristaltic pump was used, it was subject to pulsations because revolving rollers are used to compress a flexible tube with fluid inside of it, as illustrated earlier in Figure 3.2. The pulsations, however, did not interfere with the experimental pressure drops at the sampling rate used (500 samples in 0.5 seconds). This aspect is covered in more detail in section 3.4.

3.3 Pressure Transducer Calibration

The pressure transducers were calibrated by using known pressures and the measured response of the transducer. An Omega portable pressure calibrator model DPI 610 was used to supply a known pressure. The range of the pressure calibrator is

![Figure 3.4. Calibration curve for peristaltic using STA-PURE tubing.](image-url)
between -103 kPa to 206 kPa (-15 to 30 psi). The high side pressure port on the differential pressure transducer is exposed to the known value of pressure and the corresponding voltage output is read. A sample calibration curve is given in Figure 3.5 which illustrates linear behavior.

![Figure 3.5. Example calibration curve for a 103 kPa (15 psi) pressure transducer.](image)

\[ y = 1.0174x + 1.5476 \]
\[ R^2 = 1 \]

3.4 Data Acquisition

A data acquisition (DAQ) system monitors the pressure transducers in the flow loop. The DAQ is based upon the signal conditioning SCXI system from National Instruments. This system conditions all incoming signals and gives the ability to have high channel counts. The DAQ system used is capable of sampling at a rate of 100,000
samples per second. The fast sampling is used to increase the accuracy. The procedure for data collection begins with the sampling rate. The channels are sampled at a rate of 1.0 kHz. The data collected is in a waveform. The channel information contains both amplitude and frequency information. For each experimental data point 500 samples from every channel are collected. The uncertainty in the flowrate and pressure measurements are given in Table 3.1. The pressure uncertainty is larger than it might be for a gear type pump. This is because with a peristaltic pump the rollers that move the fluid create pulsations. In addition, the measured uncertainty in the flowrate tends to increase with flowrate when observing the data from Table 3.1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flowrate Set Point (ml/min) using STA-PURE Tubing</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.009 ml/min</td>
</tr>
<tr>
<td>10</td>
<td>0.01 ml/min</td>
</tr>
<tr>
<td>30</td>
<td>0.12 ml/min</td>
</tr>
<tr>
<td>45</td>
<td>0.09 ml/min</td>
</tr>
<tr>
<td>60</td>
<td>0.15 ml/min</td>
</tr>
<tr>
<td>Pressure Sensor</td>
<td></td>
</tr>
<tr>
<td>1 psi</td>
<td>0.014 kPa (0.002 psi)</td>
</tr>
<tr>
<td>15 psi</td>
<td>0.14 kPa (0.02 psi)</td>
</tr>
</tbody>
</table>

3.5 Test Section Design

The design of the test section used to conduct the experiments is discussed here. The test section is located in the flow loop schematic and provides support for the device.
In addition, it is a platform that contains the fluidic connections necessary for data acquisition.

3.51 Test Device

Much of the particulate fouling work was conducted with microchannel devices whose general form is illustrated in Figure 2.18 in Chapter 2. The dimensions of the actual microchannels are given in Figure 3.6 and Table 3.2. Throughout this document it will be referred to as the R9 chip.

![Figure 3.6. Schematic of the geometrical parameters of the R9 microchannels.](image)

Table 3.2. Channel geometries of R9 test device.

<table>
<thead>
<tr>
<th>Hydraulic Diameter (µm)</th>
<th>2b (µm)</th>
<th>2a (µm)</th>
<th>2s (µm)</th>
<th>Pitch (µm)</th>
<th>Number of Channels</th>
<th>Channel Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>225</td>
<td>249</td>
<td>205</td>
<td>97</td>
<td>302</td>
<td>26</td>
<td>10</td>
</tr>
</tbody>
</table>

Figure 3.6 shows the location of the geometric variables used in Table 3.2 where 2a is the microchannel width, 2s is the fin thickness separating the microchannels and 2b is the depth of the microchannels. The total width and length of the microchannel array are 8 and 10 mm, respectively.
The R9 microchannels were fabricated in silicon using deep reactive ion etching (DRIE) because very high aspect ratios with straight sidewalls can be achieved with this etching technique. 8-inch wafers were used in the fabrication so the unetched substrates have a thickness of 725 µm. Figure 2.18 shows an example of the silicon microchannels. There are also inlet and outlet headers formed in the silicon. The back side of the header is rounded to help direct the flow and minimize stagnation regions. Finally, the entrance to each channel is rounded to reduce the pressure drop due to entrance effects. The radius of curvature of the channels is one half the fin thickness, s. The pressure is measured in the plenums. The plenums were carefully designed to allow for sufficient volume to reduce the fluid velocity. This ensures that only the static pressure is measured since the dynamic pressure component is very small. The silicon chip is sealed with a Pyrex cover by compression to form a fully functional microchannel device. This is shown in Figure 3.7. The cover piece has the same overall dimensions as the silicon substrate. The thickness of the cover plate is 600 µm. The cover piece is laser drilled with two holes to match with the location of the inlet and outlet headers. The diameter of the plenum holes is 1.5 mm.

![Diagram of silicon microchannels with Pyrex cover plate. Together these form the complete test device.](image_url)
Two other additional channels were used in the experimental study. The second design consists of only 9 channels with a near equal fin and channel width of about 400 µm. The depth is 151 µm giving a hydraulic diameter of 220 µm which is similar to the R9 chip previously discussed. The design of the chip is given in Figure 3.8 and Table 3.3. The inlets and outlets of the channels have angles of 120°. Chips made from these designs are referred to as J2-chips. These chips were made from 6-inch silicon wafers with an unetched substrate thickness of 625 µm.

![Diagram of microchannels](image)

**Figure 3.8.** Diagram shows layout of microchannels consisting of 9 channels with inlet and exit angles of 120°.

**Table 3.3.** Channel geometries of J2 test device.

<table>
<thead>
<tr>
<th>Hydraulic Diameter (µm)</th>
<th>2b (µm)</th>
<th>2a (µm)</th>
<th>2s (µm)</th>
<th>Pitch (µm)</th>
<th>Number of Channels</th>
<th>Channel Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>220</td>
<td>151</td>
<td>407</td>
<td>393</td>
<td>800</td>
<td>9</td>
<td>10</td>
</tr>
</tbody>
</table>
The third type of microchannel device used has an offset strip fin geometry. A SEM micrograph of a similar sister chip is shown in Figure 3.9. These are often referred to as enhanced channels because in heat transfer applications, the offset fins help break up the thermal boundary layer periodically thereby providing increased heat transfer. The geometries of this microchannel are given in Table 3.4. These channels have the same unetched substrate thickness of the R9 chip which is 725 µm and possess the same overall microchannel design for the inlet and outlet headers as shown in Figure 2.18.

Figure 3.9. SEM micrograph of microchannels with offset strip fins.
Table 3.4. Channel geometries of G2-036 test device.

<table>
<thead>
<tr>
<th>Distance between fins (µm)</th>
<th>Fin thickness (µm)</th>
<th>Fin Length (µm)</th>
<th>Number of fins across channel</th>
<th>Channel Depth (µm)</th>
<th>Overall channel width (mm)</th>
<th>Overall Channel Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>40</td>
<td>500</td>
<td>47</td>
<td>200</td>
<td>8</td>
<td>10</td>
</tr>
</tbody>
</table>

3.52 Test Section

The test section was constructed with black Delrin (trade name for acetal polyoxymethylene) which is an acetal resin. It is a lightweight polymer that is very stable and is capable of being machined to fairly high tolerances. The test fixture is comprised of the following major parts: (1) main block, (2) device retaining plate and (3) mounting plate. The main block holds the test device. It is machined to allow the test device to easily rest within it and is illustrated in Figure 3.10. The entire test fixture assembly is displayed in Figure 3.11. Complete dimensioned drawings are in the Appendix section A.

A critical part of the test fixture assembly is the device retaining plate shown in Figure 3.12. It provides the clamping force on the microchannel test section and fluidic connections. This piece is made of Lexan (trade name) which is an optically clear polycarbonate material. The fluid enters the piece and moves into the inlet plenum. The fluid then enters the test device and leaves through the exit plenum.

Improved repeatability of the experiments was obtained by using vertical pressure taps on the test fixture. This was done because particles could leave the test loop by pulsations when side pressure tap lines were used. The silica and alumina particles being used were denser than water. Therefore, if they did get forced into the pressure tap lines
they would eventually settle back into the system due to gravitational effects so that the particle concentration will remain constant.

Figure 3.13 illustrates the stacked layer of materials used between the main block and device retaining plate. There are two separate thin pieces of silicone rubber membrane 200 µm thick from Acme Rubber which reside between (1) the device retaining plate and the Pyrex cover and (2) the silicon chip and the main block. The silicone rubber membranes help provide compliance during assembly. With them a good seal is provided and breaking of the test device is prevented.
Figure 3.11. Test fixture assembly.

Figure 3.12. Device retaining plate.
Figure 3.13. Side view of material stack with silicon membrane material to provide a good seal and prevent fracturing of the test device while under compression.

3.6 Glassware, Water, Commercial Reagents and Filters

All glassware used was initially cleaned with brushes and detergent to remove any visible contamination, after which it was thoroughly rinsed and allowed to dry before use. Deionized water was used which had a resistivity of 18 MΩ·cm and a pH of 5.5. The starting pH was mildly acidic due to CO₂ adsorption which produced a small amount of carbonic acid (H₂CO₃) that increased the hydrogen ion concentration. The pH was then adjusted as needed to higher or lower values by the addition of KOH (base) or HCL (acid). The mean water temperature for all experiments was 23 °C ± 1 °C.

In experimental runs using silica particles a citric acid buffer at a concentration of 3.05 mM was used to help control the pH. At this concentration, the pH would remain stable for at least 24 hours (for a 150 ml sample). When the citric acid was used the pH was always adjusted to citric acid’s first equilibrium pKa value of 3.13. At this value the buffering capacity of the citric acid was greatest. In addition, only 1.7% of the buffer
would have a charge of $2^-$, so the assumption of a single, symmetrical electrolyte would remain reasonably valid.

It is necessary to filter out fibrous material for certain experimental runs. This was done with a 0.2-µm (Acro 50, PTFE membrane) filter from Pall. 20-µm nylon mesh filters from Spectrum Laboratories were also used in some experiments in an attempt to strain out fibers that can exist within a concentrated alumina dispersion. To perform this, the nylon mesh was held in place using a stainless steel in-line filter house.

3.7 Silica and Alumina Particles

Three different types of particles were used. They are listed in Table 3.5. The 4-µm fused silica and 150-nm alumina particles came from Nanostructured & Amorphous Materials, Inc. while the 248-nm alumina particles came from Alfa Aesar. An additional particle type used was silicon from Alfa Aesar. It was not employed in fouling experiments but was used to determine the zeta potential of the silicon microchannels for different pH conditions.

<table>
<thead>
<tr>
<th>Particle Type</th>
<th>( \text{SiO}_2 )</th>
<th>( \alpha-\text{Al}_2\text{O}_3 )</th>
<th>( \gamma-\text{Al}_2\text{O}_3 )</th>
<th>( \text{Si} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Particle Size</td>
<td>4.0 µm</td>
<td>1.25 µm$^3$</td>
<td>248 nm$^4$</td>
<td>818 nm</td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>1.7 µm</td>
<td>0.087 µm</td>
<td>1.6 nm</td>
<td>91 nm</td>
</tr>
<tr>
<td>Average Density (g/cm$^3$)</td>
<td>2.40</td>
<td>3.97</td>
<td>3.97</td>
<td>NA</td>
</tr>
<tr>
<td>Percent Purity (metals basis)</td>
<td>99.94</td>
<td>99.97</td>
<td>99.97</td>
<td>98</td>
</tr>
</tbody>
</table>

3 The size of the 1.25-µm particles were obtained by dynamic light scattering. The particles, however, came from the manufacturer as a 150 nm approximate particle size (APS) powder. The discrepancy is due to the fact that the particles exist in a well clustered form when they come from the manufacturer.

4 The size of the 248-nm particles were obtained by dynamic light scattering. The particles, however, came from the manufacturer as a 3-micron APS powder. The discrepancy is based on the fact that their particle size analysis was based on a gas absorption methods and not a direct particle size measurement.
The average particle sizes were determined with particle size analyzer instrumentation discussed in Section 3.9. Making of dispersions of alumina and silicon particles were assisted by using a Branson 200 ultrasonic cleaner. The sonicator helped break up agglomerates so that all the particles within a dispersion would be well suspended. Moreover, the sonicator was also used to remove particles stuck on glassware after experimental runs.

A field emission scanning electron microscope (FE-SEM) was used to obtain morphological data on the particles used. The images shown in Figures 3.14-3.19 indicate that none of the particles were spherical in nature. However, to simplify the analysis a spherical shape was assumed for both the individual particles as well as their aggregates.

Figure 3.14. SEM image of silica particles with an average particle size of 4.0 μm.
Figure 3.15. SEM image of silica particles with an average particle size of 4.0 µm. Particle shows stress fractures with smaller particles adhering to it.

Figure 3.16. SEM image of alumina particles with an average particle size of 1.25 µm. Image shows well clustered particles.
Figure 3.17. SEM image of alumina particles with an average particle size of 1.25 µm. Individual particles appear to be fused together. This may account for the relatively large values obtained by light scattering particle size analysis.

Figure 3.18. SEM image of alumina particles with an average particle size of 248 nm. Image shows well clustered particles.
Figure 3.19. SEM image of alumina particles with an average particle size of 248 nm.

3.8 Water pH Measurements and Calibration

The pH of the water was measured using an Oakton pH 11 hand-held meter which has an accuracy of ± 0.01 pH. It was calibrated using pH buffers which were in the range of interest (1.68, 4.01, 7.00 and 10.00) that are certified traceable to NIST standard reference material. When only deionized water was used (no buffers or large amounts of acid and base) the pH electrodes will not give accurate values. This is because deionized water does not have enough ions present for the electrode to function properly. Readings tend to drift and may give conflicting results. To correct for this, the ionic strength was increased by adding a 7% KCl solution from Thermo Electron Corporation which had the trade name PHisa. 1 ml of this solution was added to every 100 ml of sample. This
increased the ionic concentration in the solution without affecting the pH so that stable and consistent readings could be obtained. The KCl solution was used to measure a sample of the deionized water which was used in the experiment. In addition, after each experimental run the KCl solution was used to confirm the value of the pH for that particular experimental run as necessary.

3.9 Particle Size Analysis

Particle size analysis was performed using a Brookhaven 90 Plus particle size analyzer which is based on light scattering theory. It was used in the analysis of the alumina and silicon particles. The 4-µm silica particles, however, were not analyzed using this instrument. They were sent to an outside laboratory for analysis which employed a disk centrifuge particle size analyzer which can accurately measure particles larger than a few microns. The report is given in the Appendix section B.

3.10 Zeta Potential Measurements

A Brookhaven 90 Plus with a BI-Zeta zeta potential analyzer was used to obtain the electrophoretic mobility of the charged particles. The temperature during operation within the measurement cell was maintained at room temperature (23 ± 1°C). When calculating zeta potentials from electromobility measurements, care must be taken to use the correct procedure based on the magnitude of $\kappa a$, where $a$ is the particle radius and $1/\kappa$ is the Debye length. An equation from Oshima (1994) was used to convert electrophoretic mobility to zeta potential which is given as

$$\zeta = \frac{3u_e \mu_f}{2ef(\kappa a)}$$  (3.1)
where \( u_E \) is the electrophoretic mobility, \( \mu \) is the dynamic viscosity, \( \varepsilon \) is the permittivity of the medium and \( f(\kappa a) \) is a monotonically varying function which increases from 1.0 at \( \kappa a = 0 \) to 1.50 at \( \kappa a = \infty \). \( f(\kappa a) \) is expressed as

\[
f(\kappa a) = 1 + \frac{1}{2\left[1 + \delta / \kappa a\right]^3}
\]

where

\[
\delta = \frac{2.5}{1 + 2e^{-\kappa a}}
\]

In the case when the particles were known to form aggregates during zeta potential measurements the average radius of the aggregate was used.

In the case of metal oxides such as \( \text{SiO}_2 \) and \( \text{Al}_2\text{O}_3 \), hydroxylated surface groups can form on contact with water, and these surface groups can ionize depending on the pH conditions as shown by Eqs. 3.4 and 3.5

\[
\text{Si} - \text{OH}^+ \overset{\text{below} \ pzc}{\leftrightarrow} \text{Si} - \text{OH} \overset{\text{above} \ pzc}{\leftrightarrow} \text{Si} - \text{O}^- \quad (3.4)
\]

\[
\text{Al} - \text{OH}^+ \overset{\text{below} \ pzc}{\leftrightarrow} \text{Al} - \text{OH} \overset{\text{above} \ pzc}{\leftrightarrow} \text{Al} - \text{O}^- \quad (3.5)
\]

The pH at which the zeta potential is zero is known as the iso-electric point or point of zero charge (pzC). At pH conditions away from the iso-electric point the net surface charge is either positive at lower pH, or negative at higher pH, reflecting the exchange of protons with the surface hydroxyl groups. This is easily seen in the above equations.

The zeta potential measurements for the particles of interest are given in Figures 3.20 – 3.22.
Figure 3.20. Zeta Potential vs. pH for silicon and 4-µm silica in a 3.05 mM citric acid solution.

Figure 3.21. Zeta potential vs. pH for silicon and 248-nm alumina particles.
Figure 3.22. Zeta potential vs. pH for silicon and 1.25-µm alumina particles.

3.11 Visualization Studies

A high magnification Keyence digital microscope, model VXH-500 was used to visualize the fouling phenomena within the microchannel test device. The microscope used a Keyence VH-Z100 wide-range zoom lens with a 10X zoom covering a 100X to 1000X magnification. The lens has a 25-mm working distance so that it can easily observe fouling behavior through the device retaining plate which is constructed from a 12.7 mm thick piece of clear polycarbonate.

The microscope is also used to capture videos of the fouling phenomena at a frame rate of 15 frames per second. In addition, time elapsed photography was employed to study the fouling over a specified period of time. Typically pictures were taken every
minute from the beginning of the run until the end. The microscope was calibrated using a standard from Bausch and Lomb for making accurate lateral measurements. The calibration factors for different magnification levels are provided in Table 3.6.

Table 3.6. Calibration factors in both the x and y directions for the Keyance VXH-500/VH-Z100 system.

<table>
<thead>
<tr>
<th>Magnification</th>
<th>Calibration Factor</th>
<th>Standard Deviation</th>
<th>% COV</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 X</td>
<td>0.9715</td>
<td>0.00135</td>
<td>0.14</td>
</tr>
<tr>
<td>200 X</td>
<td>0.9539</td>
<td>0.00034</td>
<td>0.04</td>
</tr>
<tr>
<td>300 X</td>
<td>0.9633</td>
<td>0.00344</td>
<td>0.36</td>
</tr>
<tr>
<td>500 X</td>
<td>0.9695</td>
<td>0.00000</td>
<td>0.00</td>
</tr>
<tr>
<td>800 X</td>
<td>0.9751</td>
<td>0.00265</td>
<td>0.27</td>
</tr>
<tr>
<td>1000 X</td>
<td>0.9629</td>
<td>0.00111</td>
<td>0.11</td>
</tr>
</tbody>
</table>

3.12 Micro-PIV Studies

A µPIV system from TSI Incorporated was employed in this study. The schematic of the setup is shown in Figure 3.23. The µPIV system was assembled around a Nikon Eclipse TE2000-S inverted microscope. The illumination beam was produced by a 90 mJ PIV Nd: YAG laser system from New Wave Research with a peak emission of 532 nm. The beam coming from the laser head is collimated by a set of diffusion filters to provide a uniform light source. The beam then is passed to an epi-fluorescent filter cube where part of the light is reflected upward towards a Nikon CFI Plan Fluor 10X objective lens with a numerical aperture of 0.30. The objective lens relays the light onto the microfluidic device, where it illuminates the entire flow volume. Fluorescent particles in the flow field absorb light at $\lambda = 532$ nm and emit at $\lambda = 560$ nm. The light is then imaged by the objective lens and passed back through the filter cube where it is transmitted to a

---

5 The filter cube has a coating that is designed to reflect at $\lambda = 532$ nm and transmit 560 nm.
CCD camera (630047 PIVCAM 13-8 image capture system). The images from the camera are then downloaded to a personal computer for processing. A laser pulse synchronizer (model 610034) was used to synchronize the laser and CCD camera to process the resulting image pairs so that velocity vectors and flow patterns could be ascertained. INSIGHT 3G™ software from TSI Incorporated was used to process all the files captured by the µPIV system. All experiments used an interrogation area of 128x128 pixels. Moreover, the test fixture for the µPIV studies is shown in Figures 3.24A-B. Complete dimensioned drawings are in the Appendix section A.

![Schematic diagram of the µPIV system](image)

*Figure 3.23. Schematic diagram of the µPIV system, Meinhart et al. (2000).*
Figure 3.24A. Front view of test fixture for µPIV system.

Figure 3.24B. Bottom side view of test fixture for µPIV system.
Polystyrene microspheres from Invitrogen with a diameter of 1.0 μm were used which were fluorescently tagged to excite at 540 nm and emit at 560 nm. The particles in water, however, actually swell to a size of 1.1 μm. They came from the manufacturer with a concentration of 1.0x10^{10} particles/ml and were diluted by 10:1 with deionized water when used as tracers in the μPIV work.

The fluid was run through the microchannels using a Genie Plus syringe pump from Kent Scientific. The pump was calibrated by weighing the amount of fluid collected over a given period of time for certain volumetric flowrates. This was done three times for each flowrate set point where the average and standard deviation were calculated. The calibration curve is presented in Figure 3.25 which shows a linear correlation.

![Figure 3.25. Calibration curve for syringe pump.](image-url)
A 1 µm displacement of the objective lens was determined to actually give a 1.31 µm displacement when imaging through the test section filled with latex particles in water. This is close to what would be assumed theoretically using Snell’s law when water alone is in the channels. The theoretical value can be determined from the ratio of the refractive indexes of water and air. Therefore, a 1 µm displacement of the objective lens should result in \( n_{\text{water}}/n_{\text{air}} = 1.33/1.00 = 1.33 \) µm displacement of the objective plane. The calibration of the instrument was performed by using a microscope standard from Bausch and Lomb.

The wall shear stress is given by Eq. 3.6 which is simply equal to the dynamic viscosity times the velocity gradient at the wall also known as the shear rate.

\[
\tau_w = \mu f \left( \frac{du}{dz} \right)_w
\]  

Eq. 3.6

Using this equation a graph of velocity vs. position can be constructed using the µ-PIV. The velocity gradient can easily be determined by evaluating the slope of the graph at the channel wall. Initial µ-PIV studies were performed in the microchannels of the R9 chip. This data is presented below. Figure 3.26 shows a parabolic profile for the velocity magnitude at the channel center line vs. position which is in agreement with theory. The velocity magnitude is given by Eq. 3.7

\[
V_{\text{mag}} = \sqrt{V_x^2 + V_y^2}
\]  

Eq. 3.7

where \( V_x \) and \( V_y \) are the velocities in the x and y directions, respectively. The profile was constructed by taking measurements starting from the bottom of the channel with a microscope objective and traversing upwards towards a Pyrex cover plate that enclosed the channels. The slope of the average shear rate is determined by constructing a linear
curve using data points close of the surface of interest. Data adjacent to the Pyrex surface was primarily used to obtain shear stress data since reflections at the silicon surface gave increased variance in the velocity measurements.

Figure 3.26. Velocity magnitude at channel center vs. position for R9 chip. Data was taken within a single microchannel at a flowrate of 5.93 ml/min.

Validation of the µPIV technique was performed using the R9 microchannels with a flowrate of 1.00 ml/min. The low flowrate was used because the velocity in the channels at this value has the same velocity magnitude in the header region when the microchannel test device is operating at 5.93 ml/min as used in the fouling studies. At the higher flowrate the correlation between theory and experiment was relatively poor. The experimental value was 29% less than the theoretically predicted shear rate.

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6 The 5.93 ml/min flowrate was used because fouling studies were correlated to theory using the µPIV data at this particular flowrate.
However, at the 1.00 ml/min flowrate the µPIV data furnished good results. The data is provided in Figure 3.27. The deviation in the shear rate was 4.2% less than expected. The lower empirical velocity magnitudes can be attributed to the large measurement depth of 35 µm when using the 10X objective. The large measurement depth (similar to depth of focus of a lens) can have an averaging effect on the velocity measurements and hence reduce the shear rate values (i.e., at a distance of 20 µm from the channel surface the particles-image intensity between 2.5 to 37.5 µm away from the surface can be sufficient to affect the velocity measurements). Regardless of this, the lower 10X objective was utilized because it enabled large areas in the header region of the microchannel test devices to be studied. The calculation for measurement depth, \( \delta z_m \), was derived by Meinhart et al. (2000) and is given as

\[
\delta z_m = \frac{3n\lambda_o}{NA^2} + \frac{2.16d_p}{\tan \theta} + d_p
\]

where \( n \) is the refractive index of the fluid, \( \lambda_o \) is the emitted wavelength of the fluorescent particles, \( NA \) is the numerical aperture of the objective lens, \( d_p \) is the diameter of the particles and \( \theta \) is the collection angle of the objective lens. In addition, the numerical aperture is given by the following expression

\[
NA = n \sin \theta
\]
Figure 3.27. Average velocity magnitude in a single microchannel of the R9 chip.

Data was taken at a flowrate of 1.00 ml/min.

The theoretical velocity values were obtained by employing the following equations from, Kakac (1987). Given a fully developed flow in a rectangular duct the velocity profile of velocity, \( u \), can be expressed using Eqs. 3.10 – 3.12

\[
\frac{u}{u_{\text{max}}} = \left[1 - \left(\frac{y}{b}\right)^n \right]\left[1 - \left(\frac{z}{c}\right)^m \right]
\]  

(3.10)

\[
\frac{u_{\text{max}}}{u_m} = \left(\frac{m+1}{m}\right)\left(\frac{n+1}{n}\right)
\]  

(3.11)
\[ m = 1.7 + 0.5\alpha_c^{*-1.4} \]
\[ n = 2 \quad \text{for} \quad \alpha_c^* \leq \frac{1}{3} \]  
\[ n = 2 + 0.3\left(\alpha_c^* - \frac{1}{3}\right) \quad \text{for} \quad \alpha_c^* \geq \frac{1}{3} \]  

where \( u_{\text{max}} \) is the maximum velocity at the center of the channel, \( u_m \) is the mean velocity and \( \alpha_c^* \) is the aspect ratio of the channel which must be less than unity. The dimensions for the variables \( b, c, y \) and \( z \) are provided in Figure 3.28 which possesses symmetry along the central axis. Using these equations yields velocity profiles that are within 1\% of those computed using more exact relations, Kakac (1987). With the above equations it is possible to easily determine the average velocity over the entire width of a microchannel for a given distance \( z \) from the surface. This is done by using the fact that for a function, the average value over a given length, \( L \), is expressed as

\[ \bar{U} = \frac{1}{L} \int_0^L U dy \]  

Therefore, the average velocity at a distance \( z \) from the boundary is given as

\[ \bar{U}(y) = \frac{u_{\text{max}}}{b} \int_0^b 1 - \left(\frac{y}{b}\right)^n \left[ 1 - \left(\frac{c - z}{c}\right)^m \right] dy \]  

where both \( a \) and \( b \) are half the channel width and half the channel height, respectively.
Figure 3.28. Coordinate system for rectangular duct for Eqs. 3.10 – 3.14.

With the aforementioned knowledge, with the experimental materials, flow loop, data acquisition, and analytical instrumentation, it is now possible to investigate particulate fouling within microchannel devices. The results are given in the forthcoming chapters which make reference to the information given here.
CHAPTER 4
MODEL FOR PARTICULATE FOULING IN MICROCHANNELS

4.1 Introduction

This chapter covers the multi-scale force balance model for particulate fouling within a microchannel device. It uses existing theory which was discussed in the literature review and applies it to the microchannel configuration used in the dissertation work. The model covers a scale spanning several orders of magnitude. In addition, it includes van der Waals forces, electrostatic forces, fluid-mechanics-related forces due to shear/lift, and a body force due to gravity.

4.2 Force Based Model For Microchannel Particulate Fouling

When a particle is in near contact with a horizontal channel wall, many forces around the particle are involved in determining whether or not adhesion will occur. In this model the forces are categorized into two general main groups. The first group is the adhesive forces which are due to van der Waals force, $F_{vdw}$ (Eq. 2.25), and gravity, $F_g$ (Eq. 2.47). The second group is the removal forces which include the hydrodynamic lift force, $F_L$ (Eq. 2.40), and electrostatic forces, $F_{el}$ (Eq. 2.35). Typically the electrostatic forces are adjusted to be repulsive. However, the electrostatic forces in actuality may be attractive or repulsive in nature by adjusting the pH of the aqueous medium. If it is made attractive, the sign will change for the electrostatic force in Eq. 4.1. A schematic showing
all these forces is given in Figure 4.1. The total force, $F_T$, on a particle is the sum of the adhesive and removal forces and can be expressed as

$$F_T = F_{vdw} + F_g - F_{el} - F_L$$

(4.1)

An important aspect that requires addressing is the fact this research is looking at the attachment of a particle rather than its detachment. In doing so, particle breakaway from the surface is not being studied which includes such forces as (a) the fluid drag force on the particle and the associated resistance to rolling which counteracts it, (b) the change in contact area of a deposited particle caused by particle deformation and (c) the shape of an aggregate attached to a surface.

![Figure 4.1. Schematic showing the forces on a particle in near contact with a horizontal channel wall.](image)
4.3 Force Based Model vs. Particle Size

The forced-based model given in Eq. 4.1 will be dominated by gravitational and lift forces for large particles (≈ one micron to tens of microns in diameter). This is attributed to the fact that the gravitational and lift forces are proportional to the third and forth power of the particle diameter, respectively. However, when the particle diameter is submicron in size the force balance will be dominated by the DLVO forces. Consequently, it is more insightful to employ the potential-energy diagrams which were discussed in Section 2.4.3 of the Literature Review. This allows electrostatic forces due to particle-wall interactions as well as particle-particle interactions to be investigated. By doing so, the effect of pH can be clearly observed. This is because the surfaces involved within the fouling experiments can be adjusted to have a positive, negative or neutral electrostatic charge. Consequently, by focusing on the DLVO forces, Equations 2.38A-B and 2.39A-B can be used in the overall analysis of the particulate fouling when unaggregated submicron particles are involved.

4.4 Conclusions

Conventional theories on the forces between a particle and a wall are extended to particulate fouling within a microchannel device. This includes the use of colloidal science by using potential-energy diagrams when submicron particles are involved since the DLVO forces are dominant. The model given will be compared to the experimental results in Chapters 6-7.
CHAPTER 5

MIXED FOULING PHEONMENA

5.1 Introduction

This chapter covers a mixed fouling phenomenon that was observed within the R9 microchannel device with silica dispersions at a pH = 3.13 and a citric acid concentration of 3.05 mM. Mixed fouling in this context refers to the presence of two different foulants–particulates and fibers. It occurs when fibrous elements exist within a particle dispersion. It demonstrates that the fouling behavior due to fibrous material is quite different. In fact, the presence of fibers is extremely detrimental to pressure drops within a microchannel device.

5.2 Random Behavior Due to Mixed Fouling

During some of the early experimental runs, great variability in the pressure drop data prompted an in-depth study to be performed; by doing so it allowed for the variability which was occurring to be better understood and mitigated. The fouling occurring in the R9 microchannel device had a random component to it. A relative standard deviation of 60 % in the pressure drop after a 24 hour run was not uncommon. Figure 5.1 demonstrates the gross variability in pressure drop data. The figure shows the experimental data for three runs which were conducted under exactly identical conditions on different days. In each case the device was flowing a 0.004 wt% silica dispersion at a flowrate of 51 ml/min (channel velocity of 0.64 m/s). Both runs 1 and 2 show asymptotic behavior which leveled off after about 800 minutes of operation. However, run 3 showed
virtually no increase in pressure drop even after 24 hours. It was the focus of this work to explain these types of discrepancies. When an increase in pressure drop does occur, it generally happens within the first several minutes of operation. It then levels off after about 300 minutes. This was the case for most of the work performed when using flowrates between 35 and 67 ml/min in the test device. Consequently, subsequent fouling experiments were reduced in time from 24 to 5 hours to gather more data in a given period of time.

The increase in pressure drop, under this confounding influence can be significant. Figure 5.1 demonstrates an over 200% increase in pressure drop. The increased pressure drop will occur within a relatively short period of time. Therefore, care must be taken to prevent this phenomenon from occurring in a microfluidic device because catastrophic pressure drops can occur with only a short initiation period.

Randomness is part of the fouling process, but severe fouling is not supported by the model which was presented in Chapter 4. Random fouling reported by other researchers was given in Figures 2.25 and 2.26 of the literature review. Further experimentation was therefore necessary to identify the reasons for these discrepancies. Consequently, a dedicated, high resolution microscope with high-speed imaging capability was obtained to assist in this endeavor.
Figure 5.1. Three runs for pressure drop data across microchannels manifolds for R9 chip with a 0.004 wt% silica dispersion (flowrate = 51 ml/min, channel velocity = 0.64 m/s, pH 3.12, citric acid = 3.05 mM). Peristaltic tubing was made of Norprene not STA-PURE as used in the rest of this work. In addition, the citric acid solution was not filtered to remove fibers.

5.3 Observation of Mixed Fouling

It was observed that the fouling pressure drop data being studied from early experiments was actually a mixed fouling phenomenon at the channel inlets even after improved, spallation free tubing was employed in the peristaltic pumps. The source of some of the fibrous material was found to be from the citric acid buffer solutions. Further
pressure drop data was collected with buffer solutions that had been filtered though a 0.2-µm filter to help minimize this confounding influence. Figure 5.2 shows that filtering the buffered solution before adding silica particles helped minimize the degree of fouling in the microchannels. Moreover, this figure shows an interesting trend. The pressure drop increases with flowrate in the test device. The reasoning for this will be discussed in section 5.5.

Figure 5.2. Comparison of fouling pressure drop after 5 hours for a silica concentration of 0.001 wt%. The “F” stands for a filtered buffered solution. Filtering citric acid buffer solutions helps remove fibrous material that contributes to increased pressure drops.
A picture taken with a digital microscope is shown in Figure 5.3. It shows what is actually occurring within the channels, and in particular the header region. The fibers are being caught at the channel entrances. This phenomenon commences in part because some of the fibers are several hundred microns long and come to rest at this constriction point.

Figure 5.3. Picture from digital microscope while using R9 chip for silica particulate fouling. Notice that fibers are being caught at the channel entrance which are the main contributor to pressure drops in microchannel fouling. Run 196 with a particulate concentration of 0.004 wt%.
5.4 Potential Energy Barrier to Fouling

When observing the potential energy barrier which was calculated using Eqs. 2.37 and 2.38A-B between the silica particles and the silicon microchannels it is apparent that this barrier is too large for particulate fouling to occur by simple particle/wall interactions. This is displayed in Figure 5.4. The electrostatic forces due to repulsion are well above $10 \text{kT}$. In colloidal science an energy barrier of $10 \text{kT}$ is large enough to prevent two surfaces from contacting each other while under Brownian motion.

Figure 5.4. Potential energy-distance diagram for a silica particle approaching a silicon surface. Energy barrier is well over $1000 \text{kT}$. Figure is based on a $4.0 \mu\text{m}$ silica particle approaching a silicon flat plate, Citric acid conc. = $3.05 \text{mM}$, pH = 3.13.
5.5 High Wall Shear Stresses

Insight can be gained by looking at what other researchers have done with particle adhesion studies. It has been shown by Yiantsios and Karabelas (1995) that as the flowrate or wall shear stress is increased the percent of particles adhering to a surface will decrease. Their work aids in understanding how the mixed fouling with the silica particles and, fibrous material was behaving in a manner contrary to conventional particle depositions.

When comparing data from Yiantsios and Karabelas (1995) who had looked at the adhesive strength of 5-µm glass spheres on a glass surface it is apparent that the shear stress in the microchannels being used is large enough so that particulate adhesion is extremely unlikely. Figure 5.5 shows the percent of adhering glass particles on a glass surface vs. shear stress at the wall. This graph was made by allowing particles to settle on a horizontal glass surface. The particles were allowed to remain on the surface from 1 to 8 hours. After this time, the flow was adjusted until all particles amenable to detachment at that level of hydrodynamic force were removed. Then gradually the flowrate was increased to a higher level and the procedure repeated until almost all of the particles were removed from the surface. In all experiments the surfaces involved were kept at electrostatically neutral conditions by adjusting the pH of the aqueous environment. Therefore, only gravitational and van der Waals forces along with the hydrodynamic forces were in operation during the experiments. Nearly all the particles were removed at a wall shear stress of 90 dyne/cm². In comparison with the microchannels which were being used for silica particle fouling studies the wall shear stress varied from 134 to 257
dyne/cm². The shear calculations were based on perfectly rectangular microchannels with dimensions given in Table 3.3.

Figure 5.5. Adhesive strength distribution between 5-µm glass particles and a glass surface, Yiantsios and Karabelas (1995). The pH was adjusted to be at the isoelectric point of the glass surfaces involved.

In the silica particulate fouling it is obvious that both the high wall shear stress in the microchannels and the repulsive electrostatic energy barrier shown in Figure 5.4 both contribute to the mitigation of fouling. These two aspects are decoupled in further work with alumina particles which under appropriate pH conditions can be made electrostatically attractive to the silicon microchannels.
5.6 Study of Mixed Fouling Phenomenon

It is obvious that mixed fouling occurs at the microchannel inlets. High resolution video was used to propose a theory on how this secondary fouling effect occurs. Additionally, the particle/fiber interactions which can occur at the microchannel inlets were studied. By studying the fouling occurring at the channel entrances a basic theory was developed to explain this phenomenon. This is given below.

**BASIC SEQUENCE IN FIBER/PARTICLE FOULING**

1) Fibrous material is caught at the channel entrances, due in part to some fibers which are several hundred microns long and come to rest at this constriction point.

2) Additional fiber gets caught in this “fiber web”.

3) The “fiber web” grows thicker and deeper and also incorporates particles into it causing a greater restriction to flow. Evidence of this is given in Figure 5.6.

4) For this mixed fouling phenomenon the pressure drop increases with particle concentration because the “fiber web” becomes denser and therefore a greater obstruction to the flow. This may also be due to the fact there are more particles which can potentially scrape the walls of the tubing or polycarbonate test fixture and introduce fibers into the system.

5) Fouling can increase with flowrate because more of the fiber is available to make it to the channel inlets to initiate fouling. The reasoning is that larger fibers have an increased drag force operating on them because of their larger surface area. Eq.
5.1, from Tolliver (1988) shows this where the drag force, $F_d$, on a fiber is given as

$$F_d = R_f L_f \rho_f U_{\infty}^2 C_d$$  \hspace{1cm} (5.1)

where $L_f$ is the fiber length, and $C_d$ is a drag coefficient for the particular geometry involved.

![Silica Particles](image)

**Figure 5.6.** Picture shows silica particles attached to fibrous material after an experimental run. Fibers displayed have an upper diameter of 20 µm.

The single-fiber efficiencies for a 4-µm particle interacting with a 20-µm diameter fiber are given in Table 5.1. The single-fiber efficiency equations 2.69, 2.78, 2.81, 2.84 and 2.86 were used in the computations. The calculations assume that $U_o$ is equivalent to the mean velocity (0.64 m/s) in the R9 microchannels for a flowrate of 51 ml/min in the
test device. The data show that for the conditions given, a particle is much more likely to collide with a fiber by interception or inertial impact. In addition, because the efficiency of inertial impact is dominant one should expect that a particle is more likely to collide with a fiber and possible adhere to it at higher flowrates (see Eqs. 2.80 and 2.81). This was observed experimentally in Figure 5.2.

Table 5.1. Single-fiber efficiencies for 4-µm particle and 20-µm diameter fiber. Values are for the following conditions: Fluid = water, \( U_o = 0.64 \) m/s, \( \alpha = 0.1 \), \( T = 22^\circ C \), \( R_p = 2 \) µm, \( R_f = 10 \) µm, \( \rho_p = 2200 \) kg/m³.

<table>
<thead>
<tr>
<th>Single-fiber Efficiency</th>
<th>Value</th>
<th>Percent of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Interception</td>
<td>0.060</td>
<td>42.8</td>
</tr>
<tr>
<td>Inertial Impact</td>
<td>0.080</td>
<td>57.1</td>
</tr>
<tr>
<td>Diffusion</td>
<td>( 1.25 \times 10^{-5} )</td>
<td>&lt; 0.1</td>
</tr>
<tr>
<td>Gravitational Settling</td>
<td>( 1.28 \times 10^{-3} )</td>
<td>&lt; 0.1</td>
</tr>
<tr>
<td>Total</td>
<td>0.140</td>
<td>100</td>
</tr>
</tbody>
</table>

The data in Table 5.1 are consistent with the experimental results which show that increased fouling occurs at higher flowrates. This helps elucidate what was transpiring in the microchannel test device where the pressure drop was increasing with larger flowrates. This is important because the experimental results are opposite to conventional particulate fouling theory when the fibrous elements are present. These fiber interactions which have been discussed correctly predict that fouling will increase with velocity due to particle-fiber interactions. This discovery provides new insight into the fouling mechanisms within a microchannel device. Information concerning pressure drops in the absence of (or minimal amount of) fibrous material is covered in Chapter 6. In addition, other sources of fibrous materials besides the citric acid buffer are discussed in Chapter 8.
5.7 Mixed Fouling Conclusions

The fouling behavior due to fibrous elements is quite different. The presence of fibers is extremely detrimental to pressure drops within a microchannel device. The pressure can easily increase by a factor of four. These types of increased pressure drops can occur within the first several minutes of operation. In this phenomenon fibers collect at the channel entrances of a microchannel device and obstruct the flow at this constriction point. In fact this aspect has not been reported in the microchannel literature before.

Fouling in the presence of a significant amount of fibrous material will behave in a manner opposite to simple particulate fouling as discussed previously. The pressure drop was shown to actually increase with flowrate. The reasoning for this can be explained in part because a particle is more likely to collide with a fiber and possibly adhere to it by inertial impact when the flowrate is increased under the conditions studied (see Table 5.1). The end result is a greater restriction to flow and increased pressure drop.
CHAPTER 6

ALUMINA FOULING OF AGGREGATED PARTICLES

6.1 Introduction

This chapter covers the experimental results obtained with the 1.25-µm average particle size alumina. The alumina particles with proper pH adjustment of 3.0 could be electrostatically attracted to the silicon channels of the R9 chip. This allowed the fouling mitigation due to the electrostatic forces and the shear stress at the channel walls to be decoupled since only the shear forces would be operating to minimize fouling. This was done in part because the pressure drops in the microchannels could not be quantitatively modeled since the fiber that is introduced into the experimental system is random in amount and where it accumulates.

6.2 Zeta Potential Adjustment of Alumina Particles and Silicon Channels

An operating pH of 3.0 was selected for all experiments using the nominal 1.25-µm alumina particles. At this pH the alumina particles would have a positive zeta potential while the silicon zeta potential would be negative. The zeta potential vs. pH chart for alumina and silicon particles is provided in Figure 3.58 in Chapter 3 which discusses the experimental methodology. At a pH of 3.0 the absolute difference in potential between the two surfaces was 54.3 mV. This does, however, assume the zeta potential of the silicon channels can be correctly modeled with silicon particles. This is because the silicon microchannels were formed by a deep reactive ion etching (DRIE)
process which is known to leave behind a poly-fluorinated residue after etching and is very difficult to completely remove, Moon (2006).

The zeta potential vs. pH for alumina and silicon particles given in Figure 3.58 was obtained without the use of any buffers. The zeta potential within the pH range of 2.5 and 3.5 for both particle types is for the most part steady. Therefore, a pH of 3.0 should provide a fairly constant potential difference between alumina and silicon even if the pH varies somewhat about the desired operating condition. Experimentation has shown that the pH during runs has only varied to an upper limit of 3.18 which is well within expectable limits.

6.3 Alumina Fouling in Header Regions

In visualization studies particulate fouling using alumina particles was observed. In numerous trials it was demonstrated that the shear stress at the channel walls is sufficiently large to prevent particulate deposition from occurring even when there is an electrostatic attraction between the alumina particles and the silicon microchannels. When operating at a flowrate of only 5.93 ml/min for the test device no fouling within the microchannels occurred. This flowrate is far below what is typically used in microchannels for heat dissipation. Flowrates used by Steinke (2005) ranged from 39 to 69 ml/min with the same test chip for heat transfer studies. The lack of fouling in the channels can be contrasted with the particle depositions that occurred within the inlet and outlet headers of the microchannel device. A visual comparison can be performed by looking at Figures 6.1 and 6.2.

The average size of the alumina aggregates was estimated to be $24.1 \pm 2.4 \, \mu m$. This value was determined by taking the mean diameter of the aggregates with various
geometries which deposited in the header region. This was done using the digital microscope as shown in Figure 6.3 at a magnification of 500X when the alumina deposition was light enough to distinguish alumina clusters as being separate. The reason for the particle aggregation is not clear since the electrostatic forces based on using Eqs. 2.37 and 2.39A-B give an energy barrier of over 1000 kT which should be sufficient to prevent the particles from clustering together. The mean particle size performed by dynamic light scattering was determined to be 1.25 µm. This is much larger than what would be expected since the particles from the manufacturer were stated to have an average particle size of 150 nm. The SEM images of Figure 3.52 give insight to these discrepancies. It shows an image of well clustered particles. Moreover, in Figure 3.53 it appears that the smaller particles may be fused together in some manner giving an average particle size of 1.25 µm. The particle clustering may be a result of forces which are not explained by DLVO forces.

Deviations from classic DLVO theory behavior have been attributed to a number of factors. Two such reasons include discreteness of surface charge and surface roughness, Snoswell et al. (2005). Regardless of the exact reasons behind the particle aggregation this estimated mean aggregated particle size of 24.1 µm was used to determine why the alumina aggregates were depositing in the header region of the R9 chip but not in the microchannels.

The degree of aggregate coverage in the plenum increased with time which was consistent across the entire header region. The aggregates formed only a single layer with clusters breaking off from the surface periodically. These particle deposits were observed in both the inlet and outlet header regions with a tendency for greater deposition at the
inlet. The aggregates were shown to form deposition paths which followed the natural streamlines of the fluid flow in the headers.

The reason why the 1.25-µm alumina particles agglomerated in the fouling experiments can help be understood by the fact that the particles were in the alpha form (corundum). This type of alumina is not soluble in water, strong acids or bases. This facilitated their aggregation since dispersal in the aqueous environment was not attainable while the solution was under constant shear while being stirred.

Figure 6.1. Alumina particulate deposition within channel inlet header of the R9 chip. Data is from run 213 after 3 hours. pH = 3.0, flowrate = 5.93 ml/min, Alumina particle conc. = 0.016 wt %.
Figure 6.2. Microchannels of R9 chip from run 213 after 3 hours. pH = 3.0, flowrate = 5.93 ml/min, Alumina particle conc. = 0.016 wt %.

Figure 6.3. Picture of aggregated 1.25-µm alumina particles. The mean particle aggregate size was estimated to be 24.1 µm. Magnification = 500X.
6.4 Wall Shear Stress in Header and Microchannels

The wall shear stress in the header region of the R9 chip was determined using the velocity profile vs. distance information obtained with the µPIV. The data was taken in two regions of the header. The information of the first area (region 1) is given in Figures 6.4-6.6 and that of the second area (region 2) is given in Figures 6.7-6.9. The average wall shear stress in the two header regions is listed in Table 6.1. The surface of the test chip shows significant surface roughness. It was measured to have an average roughness of 1400 Å from profilometry measurements using a P2-Tencor. In Figures 6.4 and 6.7 it is apparent the surface has suffered from significant pitting from the DRIE process used to fabricate the microchannel device at IBM.

Table 6.1. µPIV results in header regions and analytical calculations for microchannels of the R9 chip.

<table>
<thead>
<tr>
<th>Area</th>
<th>Average Wall Shear Stress (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Region 1</td>
<td>0.14</td>
</tr>
<tr>
<td>Region 2</td>
<td>0.20</td>
</tr>
<tr>
<td>Microchannels</td>
<td>2.3</td>
</tr>
</tbody>
</table>

Figures 6.5 and 6.8 show the processed data from the µPIV. The image consists of two basic areas. The first is the liquid containing, header region which shows fluorescent particles with a wide range of velocities. These values are denoted by velocity vectors of different size which are enclosed with a dotted line. Outside of this area there is no fluid. Consequently, the dotted line is placed there when processing the µPIV data to limit the region of interest (ROI). This prevents the software from attempting to construct velocity vectors outside a region with fluid flow. Similarly, Figures 6.6 and 6.9 provide the same data given, respectively, in Figures 6.5 and 6.8. However, it is only for the ROI. Length
scales are given for both the x- and y-directions and the legends can be used to quickly obtain the velocity magnitudes. The details of the μPIV experiments are available in Section 3.12 from the chapter on Experimental Methodology.

The data show a larger shear stress further away from the channel entrances (region 2) because the flow is only bounded on one side in the x-y direction. Conversely, very close to the channel inlets the flow is bounded on two sides (region 1) which gives a lower average wall shear stress. In addition, the velocity magnitude in region 1 is less because it has two significant velocity components in both the x- and y-directions where in region 2 the velocity is mainly in the y-direction (velocity magnitude is at a maximum when the flow is dominant in one direction). The average wall shear stress in the microchannels was determined analytically by using Eq. 3.6 which is the basic definition of wall shear stress in conjunction with the velocity profile Eqs. 3.10-3.14. The result was a average shear stress at the bottom channel surface of 2.3 Pa. The difference in wall shear stress is approximately 12 times greater than the larger average shear stress in region 2.
Figure 6.4 Area of header region 1 of the R9 Chip where the µPIV data of Figures 6.5 and 6.6 were taken. Note the circular surface features in the header and microchannels caused by the etching process.

Figure 6.5. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 1. The dotted line encloses the interrogation region being studied.
Figure 6.6. Area of the header region 1 where the μPIV data of Figure 6.4 was taken. Region encompasses an area which is approximately 370 µm x 280 µm based on vector positions. Figure shows velocity magnitudes at an approximate distance of 52 µm from the surface in the header region.

Figure 6.7. Area of the header region 2 of the R9 Chip where the μPIV data of Figures 6.8 and 6.9 were taken.
Figure 6.8. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 2. The dotted line encloses the interrogation region being studied.
6.5 Controlling Forces of Particle Aggregates at Header and Microchannel Surfaces

Fouling occurred within the header region but not in the channels due to the competing forces of lift and gravity. Equations 2.40 and 2.47 were used in calculating these forces. It should be restated here that the lift force is proportional to the square of the wall shear stress which is much larger in the microchannels. The results are given in Table 6.2 which makes this comparison for region 2 in the header which had the larger average wall shear stress.
Table 6.2. Gravitational and lift forces in R9 chip for 24.1 µm particle aggregates. Particle density is taken as that of the bulk value. Flowrate = 5.93 ml/min in test device.

<table>
<thead>
<tr>
<th>Area of Interest</th>
<th>Gravitational Force (N)</th>
<th>Lift Force (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Header region 2</td>
<td>2.1x10^{-10}</td>
<td>8.9x10^{-12}</td>
</tr>
<tr>
<td>Microchannels</td>
<td>2.1x10^{-10}</td>
<td>6.4x10^{-10}</td>
</tr>
</tbody>
</table>

Within the header the gravitational force dominates and is about 24 times greater than the lift force\(^7\). Therefore, the particle aggregates will be able to settle on the bottom of the header. This explains in part why fouling was always observed in the R9 silicon header and not on the Pyrex cover which enclosed this area of the microfluidic device. Conversely, within the microchannels the lift force is dominant and is approximately 3 times larger. The larger lift force prevents the particle aggregates from reaching the surface and depositing themselves.

When the electrostatic and van der Waals forces are considered their impact is minimal. This is observed in Figures 6.11 and 6.12 where the absolute values of the forces involved are considered for the situation shown in Figure 6.10. Figure 6.11 compares the forces in region 2 of the header. In this case the gravitational force is greater than all other forces until a separation distance of 41 nm. Thereafter, the aggregated particles will be tightly held at the silicon surface by the attractive electrostatic forces. Figure 6.11 compares the forces involved within the R9 microchannels. Here the lift force is larger than the other forces at play until a separation distance of 30 nm. At this distance the positive electrostatic forces will dominate. However, fouling does not occur within the channels. This is because the alumina

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\(^7\) The gravitational forces were based on the bulk density of the alumina particles. When aggregates grow there is a decrease in the aggregate density. Even if the density was reduced by a factor of 2 the gravitational force would still be 8 times greater than the lift force in the header region. Calculations were based on Eqs. 2.40 and 2.47.
aggregates do not approach close enough to the silicon surface for particle deposition to occur.

Therefore, the model which was first discussed in Chapter 4 successfully predicts that the wall stress in the microchannels is too high for fouling to occur within them. Moreover, it explains the opposite behavior where significant particle deposits occurred within the header regions of the microchannel device.

Figure 6.10. Schematic showing the forces on a particle in near contact with a horizontal channel wall as shown in Chapter 4 when the fouling model was first introduced. The drag force on the particle is not included because the author is looking at the attachment of a particle rather than its detachment.
Figure 6.11. Forces involved for different separation distances for a 24.1 µm alumina aggregated particle approaching a silicon surface in header region 2 of R9 chip. pH = 3.0, $\tau_w = 0.20$ Pa.

Figure 6.12. Forces involved for different separation distances for a 24.1 µm alumina aggregated particle approaching a silicon surface in microchannels of R9 chip. pH = 3.0, $\tau_w = 2.3$ Pa.
6.6 Filtration of Fibers in Alumina Particle Dispersion

During the previous experiments the fiber that was present in the system was still considered a confounding influence. It was desired to demonstrate that in the absence of fibers there would be no increase in pressure drop within the microchannel device. To this end, alumina particle dispersions were filtered through a 20-µm nylon mesh in an attempt to strain out the fibers while still maintaining a concentrated alumina dispersion.

To help eliminate remaining fibers in the system from a previous experimental run the test loop was run for 5 minutes at a flowrate of 67 ml/min for the test device while an inline 0.2-µm filter was in place. Therefore, the system prior to introducing a particulate dispersion was void of fibrous material.

Experiments were conducted by formulating a 0.032 wt% alumina solution. This solution was then filtered twice through a clean 20-µm nylon mesh. The final particle concentration contained a significant solids content to help demonstrate in a semi-quantitative manner that in the absence of (or minimal amount of) fibrous material there will be no increase in pressure drop. The experimental results are displayed in Figures 6.13 - 6.15. Figure 6.13 shows that there was no increase in pressure drop during 3 hour runs at a flowrate of 5.93 ml/min. In contrast, Figure 6.14 demonstrates that there was still a large amount of particulate fouling which occurred within the header region primarily because of the low shear stresses involved. However, because of the high shear stresses within the microchannels there were no particle deposits. This is seen in Figure 6.15.
Figure 6.13. Pressure drop vs. time over a 3 hour period for a filtered alumina dispersion using the R9 chip. pH = 3.0, flowrate = 5.93 ml/min.

Figure 6.14. Alumina particulate deposition within channel inlet header of R9 chip from run 222 after 3 hours. pH = 3.0, flowrate = 5.93 ml/min.
The alumina particles used for these experiments aggregated very quickly in a manner that cannot be explained by DLVO theory. Therefore, it would be helpful to obtain additional experimental results using stable alumina colloids. This aspect will be covered in the following chapter.

6.7 Conclusion on Alumina Fouling of Aggregated Particles

Fouling with larger particles (i.e., 24 µm) in typical microchannels used for heat dissipation (i.e., 249 µm x 205 µm) is extremely unlikely because of the high shear stresses involved even at a low flowrate of 5.93 ml/min in the test device. This is still the case when particles are electrostatically attracted to the microchannel surface. It is the large lift forces involved which are responsible for inhibiting particle deposition. This will occur when the particles have sufficient size for the lift force to overcome the gravitational force.
The channel header is quite susceptible to particulate fouling. This is because of the low shear stress at the wall. In contrast, it is fortuitous that the microchannels do not possess the same fouling characteristics as the headers. This is especially the case since they have the smallest geometrical constraints in a microfluidic device.

In the absence of fibers, particulate fouling will not cause an increase in pressure drop even at a flowrate of 5.93 ml/min for the microchannel geometries studied. Since most channels used for heat dissipation are run at much higher flowrates ($\approx 35$ to 67 ml/min) the probability of particulates depositing themselves in the microchannels or header region is very small. In addition, the force balance model discussed in Chapter 4 was further validated because when the fibers were removed from the system the fouling characteristics were in line with the expected results.
CHAPTER 7

FOULING WITH STABLE ALUMINA PARTICLES

7.1 Introduction

This chapter covers the experimental results obtained with the 248-nm average particle size alumina at a concentration of 0.032 wt%. The fouling behavior was observed for three regions of pH using the J2 chip and the enhanced G2-036 chip with a flowrate of 5.93 ml/min in the test devices. These include the following conditions:

(1) The alumina particles are electrostatically attracted to the silicon surface.
(2) The alumina particles experience electrostatic repulsion with the silicon surface.
(3) The alumina particles are near their isoelectric point.

When the 248-nm alumina particles form a stable colloid the gravitational and lift forces are negligible compared to the DLVO forces. They are in fact several orders of magnitude lower. This can be seen in Figure 7.0 which compares the forces between a 248-nm particle and a silicon surface for the conditions specified. Consequently, the bulk of the discussion will center on the DLVO forces. Since the DLVO forces are dominant the potential-energy diagrams are used to gain insight on particle-wall as well as particle-particle interactions. In addition, the pH was adjusted in the experiments to validate its effect for the model discussed in Chapter 4.

Unlike the alpha type particles which agglomerated in the fouling studies of Chapter 6, the 248-nm particles were of the gamma form which is also known as active
alumina. This form can more easily be dispersed and hence allowed for another aspect of alumina fouling to be studied.

![Force vs Distance](image)

**Figure 7.0** Forces involved for different separation distances for a 248-nm alumina particle approaching a silicon surface. pH = 3.0, \( \tau_w = 0.29 \) Pa. Plot displays the absolute value of the forces which are involved.

### 7.2 J2 Chip Fouling with Electrostatic Attraction between Alumina and Silicon Surfaces

At an operating pH of 3.0 the surfaces of the 248-nm alumina particles and the silicon test device are electrostatically attracted to each other with an absolute difference in zeta potential between them of 62.8 mV. This is observable in Figure 3.57 of Chapter 3
on Experimental Methodology. In addition, at this pH the alumina has a zeta potential of +40.1 mV which according to DLVO theory will give a stable colloid with an energy barrier of over 200 kT between colloidal particles. These calculations were based on Eqs. 2.37 and 2.39A-B in the Literature Review of chapter 2. This stability was observed by the fact that the particles at this pH did not aggregate together.

When running a 0.032 wt% alumina dispersion through the test device at a flowrate of 5.93 ml/min (channel velocity of 0.18 m/s) no fouling was observed. This can be seen by comparing Figures 7.1 and 7.2. Figure 7.1 shows the header of the J2 chip prior to the experimental run while Figure 7.2 shows the lack of particle deposition after 3 hours. At first glance this may seem odd since the particles are highly attracted to the silicon surface. However, it is well known that stable colloidal suspensions produce particle monolayers only, while further deposition is inhibited by unfavorable physicochemical interactions between deposited and suspended particles, Yiantsios et al. (1999). Several experiments have been performed with colloidal particles such as latex and silica which support this conclusion. Bowen and Epstein (1979) found in their studies that the maximum coverage never exceeded 10 % for silica particles between 400 - 650 nm in size. In addition, Dabroś et al. (1983) discovered that a single 500 nm polystyrene sphere was able to block (other identical polystyrene spheres) for an area of about 20 to 30 times its geometrical cross-section. Experimental results will of course vary depending on conditions like double layer thickness, particle size and flowrate.
Figure 7.1 Picture of J2 header region at start of run 247. 0.032 wt% alumina, pH\text{start} = 3.0, flowrate = 5.93 ml/min. Magnification = 100X.

Figure 7.2. Absence of alumina particulate deposition within channel inlet header of J2 chip. From run 247 after 3 hours. 0.032 wt% alumina, pH\text{start} = 3.0, flowrate = 5.93 ml/min. Magnification = 100X.
7.3 J2 Chip Fouling with Electrostatic Repulsion between Alumina and Silicon Surfaces

At a pH of 10.0 and 11.0 the zeta potential for the 248-nm alumina particles as shown in Figure 3.57 had a repulsive force between them and the silicon surfaces of the J2 chip. At the lower pH the alumina particles themselves had a zeta potential of -15.3 mV which gave them an energy barrier of 33 kT between particles which was enough for the colloid to remain stable. Refer to Figure 7.3 for the potential energy-distance curves which were made by using Eqs. 2.37 and 2.39A-B. An even greater stability was obtained when operating at a pH of 11.0 which had a zeta potential of -28.7 mV and an energy barrier of 113 kT between particles. This difference in the energy barrier was readily apparent. At a pH of 11.0 the stock solution with an alumina concentration of over 0.048 wt % would easily disperse after a couple minutes in the sonicator. However, at the lower pH of 10.0 it could take up to 10 minutes to obtain a stable colloid for use in the test loop. In addition, the gamma form of alumina is known to readily dissolve in strong acids and bases. This is in line with what was observed when the alumina particles were measured for zeta potentials. However because of this fact, experimental runs focused on pH values up to an upper limit of 10.0.

At the lower pH of 10.0 the electrostatic energy barrier between the alumina particles and the silicon test chip was 94 kT. This was determined using Eqs. 2.37 and 2.38A-B. This is sufficient to prevent fouling. However, even if fouling did occur, it would not be possible to perceive the deposited particles because only a monolayer would be formed on the silicon surfaces.
Figure 7.3 Potential energy-distance plots for 248-nm alumina particles at different pH values. Plots are only valid up to a separation distance of 13 nm because of the limitations imposed by Derjaguin’s approximation in the equations used.

The pH was not raised above a value of 11.0. This is because the high OH⁻ concentration can etch away at the silicon surfaces. During zeta potential analysis of the silicon particles it was observed that between a pH of 10.5-11.0 the silicon particles reacted with the high concentration of KOH to form a silonal such as Si(OH)₆²⁻, Seidel (1990). This caused the solution to go from slightly turbid to clear. The reason why this readily occurred after only several hours or several minutes if the colloid was sonicated is because the silicon particles have a high surface area to volume ratio.
7.4 J2 Chip Fouling Near the Isoelectric Point of the Alumina Particles

At an operating pH of 9.0-9.5, fouling within the header and channels was observed. The reasoning for this is that when operating close to the isoelectric point the alumina colloid would become unstable and start to aggregate forming very large clusters. At this starting pH the energy barrier for particle-particle interactions was lowered to about 2.0 kT or 13 kT for the pH values of 9.0 and 9.5 respectively. This lowered energy barrier which is displayed in Figure 7.3 is enough for the individual alumina particles at a pH of 9.0 to start forming doublets. The energy barrier of 13 kT may seem high enough to prevent alumina particles from aggregating (if only Brownian motion is considered). However, the alumina colloid was constantly stirred throughout the experiment which can cause orthokinetic aggregation.

Since no buffers were used to control the pH it could drift to exist at the isoelectric point of the alumina particles during an experimental run or even obtain a charge opposite to the silicon surfaces. This would leave no repulsive electrostatic forces operating between the alumina and silicon surfaces of the test device. Buffers were not used so that the electric double layer forces could be properly modeled since only single symmetrical electrolytes would be present.

The average shear stress in the header ranged from 0.29 to 1.30 Pa for regions 1 and 2, respectively, of the J2 chip. These regions are shown in Figure 7.4. The corresponding μPIV image files and colored vector plots are shown in Figures 7.5-7.6 for

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8 An approximate value for the energy barrier is given because at this pH the scaled separation distance, κa, for the 248-nm particles is only 1.04 at pH = 9.0 and 1.69 at pH = 9.5. At such a low value of κa the curvature of the surface elements will become important between the two interacting particles and Derjaguin’s approximation looses its accuracy.

9 Orthokinetic aggregation is when a colloid is agitated causing an increase in particle-particle interactions leading to aggregate formation.
region 1 and Figures 7.7-7.8 for region 2. The shear stress in the header close to the channel inlets (region 1) is much higher than what was observed for the R9 chip discussed in Chapter 5. This is because the larger channel widths provide a smaller restriction to flow. Conversely, the shear stress in the back of the header (region 2) is similar to what was obtained for the R9 chip. Yet it is slightly larger since the channel depth is less which causes an increase in the velocity (so that the law of conservation of mass will be obeyed).

Calculations could be discussed concerning the lift and gravitational forces which were at play. However, because the experiments were performed near the isoelectric point there were some confounding influences. Severe particle aggregation was present. This gave a large range in the aggregated particle size. In addition, the pH shifted to exist at the isoelectric point or past it giving a positive charge on the alumina particles. Consequently, no absolute force balance could be obtained.

Figure 7.4. Area of the header regions 1 and 2 of the J2 Chip where the µPIV data was taken.
Figure 7.5. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 1 of the J2 chip. The dotted line encloses the interrogation region being studied.

Figure 7.6 Area of the header region 1 of the J2 chip where the µPIV data of Figure 7.5 was taken. Region encompasses an area which is approximately 420 µm x 280 µm based on vector positions. Figure shows velocity magnitudes at an approximate distance of 33 µm from the surface in the header region.
Figure 7.7. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 2 of the J2 chip. The dotted line encloses the interrogation region being studied.

Figure 7.8 Area of the header region 2 of the J2 chip where the µPIV data of Figure 7.7 was taken. Region encompasses an area which is approximately 370 µm x 300 µm based on vector positions. Figure shows velocity magnitudes at an approximate distance of 33 µm from the surface in the header region.
The particulate fouling in the header of the J2 chip is demonstrated by comparing Figure 7.9 with Figures 7.10-7.11. The latter figures show that the particles which were deposited were indeed small. When comparing Figures 7.10 and 7.11 it is apparent that the particle deposits are small enough that they cannot be imaged with the digital microscope at a magnification of 300X (the same was true for similar experiments at a magnification of 500X). In addition, at a magnification of 500X, the CCD pixel size was 0.036 microns for a 1600x1200 pixel image size within the fouled header region.

![Figure 7.9. Header of J2 chip prior to experimental run 255.](image)
Figure 7.10. Alumina particulate deposition within channel inlet header of J2 chip from run 255 after 3 hours. 0.032 wt% alumina, pH_{start} = 9.5, flowrate = 5.93 ml/min. Magnification = 100X.

Figure 7.11. Alumina particulate deposition within channel inlet header of J2 chip from run 254 after 3 hours. 0.032 wt% alumina, pH_{start} = 9.5, flowrate = 5.93 ml/min. Magnification = 300X. There are some larger particles in the background which settled after the flow loop was stopped.
It should be mentioned why the J2 chip was used to study the fouling phenomenon instead of the R9 chip with the 248-nm alumina particles. The reason was that the R9 chip has a larger average surface roughness of 1400 Å which made it difficult to observe the particle depositions with the digital microscope. Conversely, the J2 chip has a much lower surface roughness of 21 Å which did not mask the particle deposits which was necessary for good visualization studies.

7.5 G2-036 Chip Fouling with Electrostatic Attraction between Alumina and Silicon Surfaces

The fouling behavior of the alumina particles in the enhanced channels was the same as that found with the conventional J2 chip for a pH of 3.0. There were no visible particle deposits in the header region or the microchannels. This can be observed by comparing Figures 7.12 and 7.13. Figure 7.12 shows the header of the J2 chip prior to the experimental run while Figure 7.13 shows the lack of particle deposition after 3 hours. As discussed previously this is due to the formation of a monolayer which inhibits further particle deposition from the stable alumina colloid. This is an encouraging result since the fouling behavior is not limited to the narrow 60 µm channels or the high alumina concentration of 0.032 wt%. The colloid stability was the primary reason for fouling mitigation even under unfavorable electrostatic conditions between the alumina particles and the silicon channels.
Figure 7.12. Picture of G2-036 header region at start of run 259. 0.032 wt% alumina, $\text{pH}_{\text{start}} = 3.0$, flowrate = 5.93 ml/min. Magnification = 100X.

Figure 7.13. Absence of alumina particulate deposition within channel inlet header of G2-036 chip. From run 259 after 3 hours. 0.032 wt% alumina, $\text{pH}_{\text{start}} = 3.0$, flowrate = 5.93 ml/min. Magnification = 100X. Note the debris within the header region is from fibrous material which was collected at the front of the channels.
7.6 G2-036 Chip Fouling with Electrostatic Repulsion between Alumina and Silicon Surfaces

As expected the fouling behavior of the alumina particles in the enhanced channels was analogous to the J2 chip for a pH of 11.0. The lack of fouling can be attributed to the stable colloid in addition to the presence of repulsive electrostatic forces that existed between the alumina particles and the silicon surfaces of the test chip.

7.7 G2-036 Chip Fouling Near the Isoelectric Point of the Alumina Particles

The fouling within the header of the G2-036 chip was more severe for an operating pH of 9.0 when compared to the conventional J2 chip. This comparison can be made by looking at Figures 7.10 and 7.14. The large build up of particle deposits at the channel entrances is easily seen in Figure 7.14. This is due in part because the fibrous material that exists within the system comes to rest at this constriction point. This is shown in Figure 7.9 for a stable alumina colloid with a pH of 11.0. However, the main reason for the significant fouling is because the colloid was destabilized by adjusting the pH to be near the isoelectric point of the alumina particles. This caused them to aggregate together. The fouling became so severe in one of the runs that a large backpressure was built up in the system. This caused the peristaltic tubing to come loose from the barbed fitting which connected the peristaltic tubing and the Chemfluor PTFE tubing.
Figure 7.14. Alumina particulate deposition within channel inlet header of G2-036 chip from run 263 after 3 hours. 0.032 wt% alumina, pH$_{\text{start}}$ = 9.0, flowrate = 5.93 ml/min. Magnification = 100X.

Figure 7.15. Lack of alumina particulate deposition within channel inlet header of G2-036 chip from run 261 after 3 hours. 0.032 wt% alumina, pH$_{\text{start}}$ = 11.0, flowrate = 5.93 ml/min. Magnification = 100X.
The average shear stress in the header ranged from 0.043 to 0.065 Pa for regions 1 and 2, respectively, of the G2-036 chip. The shear stress values were determined by the methods discussed in Chapter 3 on Experimental Methodology. These regions are shown in Figure 7.16. The corresponding µPIV images files and colored vector plots are shown in Figures 7.17-7.18 for region 1 and Figures 7.19-7.20 for region 2. The shear stress is much lower in the header region compared to the J2 and R9 chips (R9 chip was discussed in Chapter 5). This is a result of the increased flow resistance caused by the many offset strip fin structures at the end of the header (front of the channels). At a pH of 9.0 when the colloid is unstable the increased flow resistance can have a significant effect on the degree of fouling in the header region.

Figure 7.16 Area of the header regions 1 and 2 of the G2-036 chip where the µPIV data was taken.
Figure 7.17. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 1 of the G2-036 chip. The dotted line encloses the interrogation region being studied.

Figure 7.18 Area of the header region 1 of the G2-036 chip where the µPIV data of Figure 7.17 was taken. Region encompasses an area which is approximately 390 µm x 280 µm based on vector positions. Figure shows velocity magnitudes at an approximate distance of 39 µm from the surface in the header region.
Figure 7.19. µPIV image file showing fluorescent particles with a range of velocities which are denoted by velocity vectors in region 2 of the G2-036 chip. The dotted line encloses the interrogation region being studied.

Figure 7.20 Area of the header region 2 of the G2-036 chip where the µPIV data of Figure 7.19 was taken. Region encompasses an area which is approximately 370 µm x 300 µm based on vector positions. Figure shows velocity magnitudes at an approximate distance of 33 µm from the surface in the header region.
7.8 Conclusions on Fouling with Stable Alumina Particles

Even when there is a large concentration of particulates flowing through a microchannel device, a high electrostatic energy barrier between the particles themselves will prevent fouling. This is true for particles in the nanometer regime (i.e., 248-nm). In addition, a large energy barrier between the particles and the microfluidic surfaces will help prevent fouling as long as the particles have good colloidal stability. This is in line with the microchannel particulate fouling model given in Chapter 4. As mentioned previously in Chapter 4 when the particle is submicron in size, the gravitational and lift forces are negligible. In this case the model can be simplified to use only the DLVO forces. In doing so, additional information was obtained because potential-energy diagrams could be employed. This allowed particle-particle interactions to be studied in addition to particle-surface interactions. This information was used to successfully explain the experimental results.

Proper pH adjustment is critical to prevent fouling in a microchannel test device. If the particles exist close to their isoelectric point severe fouling can occur. However, care must be taken not to make the aqueous medium too corrosive (basic). This is because the high concentration of OH\(^-\) ions will etch away at the silicon surfaces over time, Seidel (1990).

Microchannels employing offset fin strips with the dimensions studied for increased heat transfer are not prone to increased particulate fouling as long as the pH is properly adjusted to prevent particle deposition. However, the inlet header region for this type of channel configuration will have a much lower velocity and wall shear stress because of the increased flow resistance at the channel inlets. Consequently, fouling in
the header area will be worse for unfavorable electrostatic conditions for a given volumetric flowrate that cannot provide enough shear at the header surfaces.

When performing fouling studies on silicon substrates it is advantageous to have a low surface roughness so that it does not unintentionally mask particles that have deposited. Consequently, care must be taken during the silicon etch process to prevent increased surface roughness from occurring. Moreover, a smoother surface should aid in the removal of a particle once it has been deposited, Hubbe (1984).
CHAPTER 8

ORIGINS OF FIBROUS MATERIAL

8.1 Introduction

This short chapter covers experimental work to discover where the fibrous material was coming from that was involved in the mixed fouling phenomenon. It is important because the fibers were responsible for increased pressure drops within the R9 microchannel device of Chapter 5. Channels that use offset fin strips are more prone to increased pressure drops because their channel dimensions are smaller. This prevents fibrous material from making it through the fin structures and causes a greater obstruction to the flow.

The work that is discussed here is in addition to what was presented earlier in Chapter 5. In the previous work, the fibrous material was a byproduct in the manufacturing of the citric acid used to control the pH. In this case the fibers are due to an interaction between the particles and the test setup.

8.2 Deionized Water

To ensure that the experimental flow loop was not responsible for producing fibrous material, deionized water was run through the system using the G2-036 chip. This chip was selected because it was effective in removing fibers from the flow loop. This is a result of the small separation distance of only 60 µm between fins. Any fiber that is in the system will eventually come to rest at the front of the channels.
To help eliminate fiber that may remain in the system from a previous experiment, the flow loop was cleaned prior to use. The test loop was first run for five minutes at a flowrate of 67 ml/min while an inline 0.2-µm filter was in place. Afterwards, deionized water was run through the test loop for three hours at a flowrate of 5.93 ml/min as used in the alumina fouling studies. Afterwards, no fibrous material was present at the channel entrances. Therefore, the test loop by itself was considered not responsible for the fibers observed during particulate fouling studies.

**8.3 Colloid of Concentrated Particulates**

The next step in the process was to use a stable 248-nm alumina colloid with a concentration of 0.032 wt % and a pH of 11.0. The same colloid was run through the G2-036 chip three consecutive times at a flowrate of 5.93 ml/min for three hours. Before each run the system was cleaned of any fibrous material by using the procedure discussed in section 8.1. The end result after each run was a large amount of fibrous material at the channel entrances. This is illustrated in Figure 8.1.

If the fibers were originating from the particles they would have been filtered out after the first or even the second run. However, the fibrous material continued to be present after each run. Therefore, it can be reasonably concluded that particles were scraping the walls of the test setup and producing the undesired fibers. Their origin may be the PTFE tubing or the polycarbonate device retaining plate. Chemical analysis was not possible since enough material was not able to be collected for test. Figure 8.2A-C shows that the fibers in this case were semitransparent. This is consistent with the proposed origin of fibers. Consequently, it should be advantages to use metal tubing in
conjunction with a glass device retaining plate in the flow loop when studying particulate fouling.

![Image of fibrous material at the front of the G2-036 chip after a three hour run.]

**Figure 8.1** Fibrous material at the front of the G2-036 chip after a three hour run.

Run 261B, 0.032 wt% 248-nm alumina, pH = 11.0, flowrate = 5.93 ml/min.

![Image of fibrous material in header of G2-036 chip after run 261B.]

**Figure 8.2A.** Fibrous material in header of G2-036 chip after run 261B.

Magnification = 100X. Fibers are about 15 – 38 µm in diameter.
Figure 8.2B. Fibrous material in header of G2-036 chip after run 261B.

Magnification = 300X. Fibers are about 15 – 38 µm in diameter.

Figure 8.2C. Fibrous material in header of G2-036 chip after run 261B.

Magnification = 500X. Fibers are about 15 – 38 µm in diameter.
8.4 Conclusions on Origins of Fibrous Material

When performing particulate fouling studies care must be taken to exclude fibrous material from entering the microfluidic device. These fibers can easily cause an obstruction to the flow because they will come to rest at the microchannel inlets. Moreover, their ability to be captured at the channel inlet increases as the channel dimensions decrease. They can be mitigated by attaching an online filter to the system to remove fibers that exist in the flow loop prior to test.

Material selection for the experimental test setup is critical. It is best to perform preliminary studies on all fibrous material that may be present. In these experiments it would have beneficial to use metal tubing in conjunction with a glass device retaining plate in the flow loop when studying particulate fouling. The added cost of the setup would reap dividends in the experimental results. The above changes to the flow loop will help minimize the presence of fibrous material which is created by particles scraping the walls of the tubing or test fixture.
CHAPTER 9

CONCLUSIONS AND FUTURE WORK

9.1 Introduction

This chapter consists of two parts. The first section summarizes the results and accomplishment of this work. Afterwards, the second half of the chapter discusses future work which could be performed to extend the research on particulate fouling in microchannel devices.

9.2 Conclusions

An experimental investigation and theoretical analysis was undertaken to study the fouling in silicon microchannels used in IC chip cooling applications. The work focused on studying the characteristics of fouling and ways to mitigate its detrimental effects. The theoretical analysis indicated that the electrostatic and shear forces were the dominant ones in the microchannel flow applications. Their magnitudes were strongly dependent on the particle diameter.

A number of experiments in particulate fouling with microchannel test devices/structures used for IC chip cooling were performed. The channels used had the smallest hydraulic diameter ever reported for particulate fouling studies. In all cases the working fluid was water. The particulates used ranged from the colloidal size range up to tens of microns in size (for particle aggregates). By having a range in particle size the physicochemical interactions as well as the hydrodynamic forces were varied which gave different fouling characteristics. In addition, the pH was varied to adjust the electrostatic
forces between the particles and the surfaces of the microchannel devices as well as the electrostatic forces of repulsion which exist between similarly charged particles. The following conclusions can be drawn from the present work. They are given below in bullet format.

- Fouling with large aggregated particles (i.e., 24 µm) in typical, rectangular, microchannels used for heat dissipation (i.e., between 200-300 µm per side) is extremely unlikely because of the high shear stresses involved even at a low flowrate of 5.93 ml/min (velocity of 0.64 m/s in channels). This is the still the case when particles are electrostatically attracted to the microchannel surface. It is the large lift forces involved which are responsible for inhibiting particle deposition. This will occur when the particles have sufficient size for the lift force to overcome the gravitational force.

- The channel header is quite susceptible to particulate fouling. This is because of the low shear stress at the wall in the header region. In contrast, it is fortuitous that the microchannels do not possess the same fouling characteristics as the headers. This is especially the case since they have the smallest geometrical constraints in a microfluidic device.

- Even when there is a large concentration of particulates flowing through a microchannel device, a high electrostatic energy barrier between the particles themselves will prevent fouling. This is true for particles in the nanometer regime
(i.e., 248 nm). In addition, a large energy barrier between the particles and the microfluidic surfaces will help prevent fouling as long as the particles have good colloidal stability.

- Proper pH adjustment is critical to prevent fouling in a microchannel test device. If the particles exist close to their isoelectric point severe fouling can occur. However, care must be taken not to make the aqueous medium too corrosive (basic). This is because the high concentration of OH⁻ ions will etch away at the silicon surfaces over time. This is supported by the thesis work and the experiments of Seidel (1990).

- When performing fouling studies on silicon substrates it is advantageous to have a low surface roughness so that it does not unintentionally mask particles that have deposited. Consequently, care must be taken during the silicon etch process to prevent increased surface roughness from occurring. Moreover, a smoother surface should aid in the removal of a particle once it has been deposited, as indicated by Hubbe (1984).

- Material selection for the experimental test setup is critical. It is best to perform preliminary studies on all fibrous material that may be present. In these experiments it would have beneficial to use metal tubing in conjunction with a glass device retaining plate in the flow loop when studying particulate fouling. The added cost of the setup would reap dividends in the experimental results. The
above changes to the flow loop will help minimize the presence of fibrous material which is created by particles scraping the walls of the tubing or test fixture.

- The headers in a microfluidic device should be well rounded and designed so that there are no stagnation regions or areas where the flow will make abrupt changes in direction. A large change in the velocity in the forward direction of flow, to a lateral one will reduce the velocity magnitude as well as the local wall shear stress.

- The fouling behavior due to fibrous elements is quite different. The presence of fibers is extremely detrimental to pressure drops within a microchannel device. Fibers collect at the channel entrances of a microchannel device and obstruct the flow at this constriction point. Moreover, the increase in pressure drop caused by fibers can occur within the first several minutes of operation. This aspect has not been reported in the microchannel literature before.

- Fouling in the presence of a significant amount of fibrous material will behave in a manner opposite to particulate fouling. The pressure drop was shown to actually increase with flowrate. The reasoning for this can be explained in part because a particle is more likely to collide with a fiber and possibly adhere to it by inertial impact when the flowrate is increased under the conditions studied (see Table 5.1). The end result is a greater restriction to flow and increased pressure drop.
• In the absence of fibers, particulate fouling will not cause an increase in pressure drop even at a flowrate of 5.93 ml/min in the microchannel geometries studied. Moreover, since most channels used for heat dissipation are run at much higher flowrates (≈ 35 to 67 ml/min) the probability of particulates depositing themselves in the microchannels or header region is very low.

• Microchannels employing offset strip fins with the dimensions studied for increased heat transfer are not prone to increased particulate fouling as long as the pH is properly adjusted to prevent particle deposition. However, the inlet header region for this type of channel configuration will have a much lower velocity or wall shear stress because of the increased flow resistance at the channel inlets. Consequently, fouling in the header area will be worse for unfavorable electrostatic conditions for a given volumetric flowrate that cannot provide enough shear at the header surfaces. Therefore, header design becomes even more important in this case.

• A multi-scale force based model was developed to help predict particle-surface and particle-particle interactions encountered in microchannel fouling. The theoretical predictions for effect of wall shear stress and effect of pH have been verified experimentally. The model provides a new way to control the fouling behavior within microchannel devices by adjusting the pH of the solution.
9.3 Future Work

The thesis work discussed in this body of work can be continued to acquire additional knowledge in the area of particulate fouling within microchannel devices. One aspect that immediately comes to the forefront based on the experimental work is the microchannel designs necessary to mitigate fouling. This is the case since any microchannel design used for IC chip cooling requires a header to adequately distribute the coolant to a number of microchannels with a larger surface to volume ratio for increased heat dissipation. However, the header necessary to feed these individual channels has been determined to be more susceptible to fouling and therefore presents additional risks associated with unwanted particle deposits. Consequently, the header is where the focus should be on future fouling studies not the microchannels. Various geometries for the header design should be explored to do this. As part of the header designs, the surface roughness needs to be effectively controlled in the DRIE process. This will help obtain consistent and meaningful results since it can have significant effects on fouling experiments.

The experimental evidence suggests that if a microfluidic device has a large header feeding a number of smaller microchannels, it may be beneficial to etch the header to a smaller depth than the channels. This will allow for an increased velocity which will translate into a larger shear stress on the surfaces of the header. This will in turn help to mitigate particulate fouling. This may be done by using a technique called one-step two level etching (OSTLE), Grande et al. (1987). With this technique, the mask used for the channels would be made of a material which erodes at a faster rate than the
masking layer used for the header. This would be especially helpful when higher flowrates cannot be used in the device.

In this dissertation work it was concluded that the pH has a significant effect on controlling particle-particle interactions as well as particle-surface interactions. However, it may not be always desirable to adjust the pH to mitigate fouling because at very high or low pH values the medium can be corrosive. Consequently, it is desirable to integrate other technologies into a microchannel device to help mitigate particulate fouling. These include but are not limited to: (1) Surface modification to give hydrophobic and hydrophilic conditions which can mitigate fouling for certain foulants, and (2) The use of surfactants to reduce the van der Waals adhesive forces between various surfaces.

It would also be beneficial to study particulate fouling at elevated temperatures and/or under a temperature gradient. This would allow for the effect of temperature on particle deposits to be observed. This can include such aspects as (1) Change in zeta potentials that affect particle-surface and particle-particle interactions, (2) The tenacity of a particle to stick to a surface or to another particle, and (3) The effect of particle deposits which provide a resistance to heat transfer within a microchannel device.


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APPENDIX

This appendix presents the details of items that were not included in the body of the paper. However, it is important to include these aspects as a matter of record. It includes the details of the 4-μm silica particle analysis which was sent out for analysis as well as CAD drawings of the test fixture components for both the flow loop and µPIV. It also includes the justification in using the expression of lift force by Leighton and Acrivos (1985) for small separations distances between a particle and a wall.

A. CAD Drawings of Fixtures

The following scaled drawings are presented on the next 9 pages.

1. Main block
2. Device retaining plate
3. Micro-PIV fixture
2x Ø3.18 ø 1.53
Need to connect to conduit coming in from side

6x Ø0.166 inch THRU
Need to clear holes for M4 screws

SCALE 0.044

R.I.T
Device Retaining Plate
Top View I

JEFFREY PERRY
TOL ± 0.025 mm

Unless otherwise noted all values are in mm

SCALE: 0.029  TYPE: PART  NAME: DEVICE_RETAINING_PLATE_01  SIZE: A  SHEET 1 OF 2
R.I.T
THERMAL ANALYSIS AND
MICROFLUIDICS LABORATORY

JEFFREY PERRY

Bottom Plate for μ-PLV System
Screw Holes for Sealing Fixture

TOL ± 0.0254 mm

All dimensions are in mm unless specified otherwise.
Edge gaps have a radius of 0.9 mm which are centered 0.9 mm away from edge.
R.I.T
THERMAL ANALYSIS AND MICROFLUIDICS LABORATORY

JEFFREY PERRY

Bottom Plate for μ-PIV System
Fluid connections on back side of plate

TOL ± 0.0254 mm

Dimensions are in mm unless specified otherwise

SCALE 1.100
B. 4-µm Silica Particle Size Analysis

The details of the particle size analysis for the 4-µm silica particles is provided on the next 2 pages.
Sample I.D.     SiO2 Particles (Run 1)
Operator I.D.   W.Bernt
Notes           powder dispersed into MeOH -> H2O added 1:3 ratio
C. Lift Force on Particle at a Small Distance from Wall

The hydrodynamic lift force between a particle and a wall was modeled with the simple analytical equation by Leighton and Acrivos (1985). This is valid when the particle is in contact with the wall. If there is a small separation between the two surfaces involved the lift force can vary with distance. However, it is a negligible amount. A more accurate expression by Cherukat and McLaughlin (1994) can be used to obtain a better estimate of the lift force. The equation for the lift force, $F_L$, is given below. It has the same form as that given by Leighton and Acrivos.

$$F_L = \frac{I \rho_f \left( \frac{\tau_w}{\mu_f} \right)^2}{16} d_p^4$$  \hspace{1cm} (C.1)

$$I = \frac{3.2397}{k^2} \left( 1.7716 + 0.2160k - 0.7292k^2 + 0.4854k^3 \right) \Lambda_g + \left( 2.0069 + 1.0575k - 2.4007k^2 + 1.3174k^3 \right) \Lambda_{G}^2$$

$$\Lambda_g = -1$$

In Eq. C.1, $I$ is a constant which can have a minimum value of 9.28. This is for a sphere in contact with the wall. The expression is for a non-rotating sphere where the velocity gradient and slip velocity approaches zero close to the wall. $\rho_f$ is the density of the fluid, $\tau_w$ is the shear stress at the wall, $\mu_f$ is the viscosity of the fluid and $d_p$ is the particle diameter. The parameter $k$ is defined as

$$k = \frac{d_p}{2L}$$  \hspace{1cm} (C.2)

where $L$ is the distance between the wall and the centre of the sphere.
Figures 6.11 and 7.0 have been replotted in Figures C.1 and C.2, respectively, using the expression by Cherukat and McLaughlin. Concerning a large aggregate of 24.1 microns the lift force only sees a small increase of 0.3% within a separation distance of 100 nm (see Figure C.1). Similarly, for a small colloidal particle of 248 nm the lift force changes by only 0.3% for a separation distance of 88 nm. Even if the separation distance is increased further the magnitude of the lift force compared to the DLVO forces is several orders of magnitude lower. Therefore, employing the simple expression of Leighton and Acrivos is justified.

![Figure C.1. Forces involved for different separation distances for a 24.1 µm alumina aggregated particle approaching a silicon surface in header region 2 of R9 chip. pH = 3.0, τ_w = 0.20 Pa. The change in lift force with distance is negligible since the large aggregate is at a separation distance of less than 100 nm (0.3 % increase).]
Figure C.2 Forces involved for different separation distances for a 248 nm alumina particle approaching a silicon surface. pH = 3.0, $\tau_w = 0.29$ Pa. Plot displays the absolute value of the forces which are involved. The change in lift force with distance is negligible up until a separation distance of 88 nm (0.3 % increase). At a distance of 100 nm the lift force is 26% greater than at the wall.