SOLID MECHANICS IN COLLOIDAL AND BACTERIAL FILTRATION

A Dissertation Presented

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ABSTRACT

Microbial and particle transportation and adhesion in porous media in electrolytic solutions is fundamental in many fields related to water treatment, drug delivery and human health. This is a complex multi-physics process which includes, but not limited to, fluid mechanics, microbial properties, surface adhesion and aqueous environments. The conventional, empirically-driven approach is based on a flow-through sand-packed column test. Although it is widely applied to predict filtration efficiency, the fundamental science and contribution of individual factors in this problem are still missing—for example, flow condition, salt concentration and microbial properties.

This dissertation addresses this problem by developing a new microfluidic test method and a theoretical model based on contact mechanics. The new microfluidic test simplifies the physics behind microbial/colloidal filtration, and allows us to focus on the surface attachment/detachment due to inter-surface and hydrodynamic interactions. Filtration efficacy was directly measured using this new microfluidic test. The results of the tests were then compared with the conventional flow-through column test. The two show a good agreement with each other. This implies that surface adsorption is the dominant filtration mechanism in the column test. The coupled effect of flow rate, salt concentration and microbial properties were analysed using the classical DLVO theory, fluid mechanics and contact mechanics. The fundamental physics that control the microbial/colloidal attachment were suggested.

We further developed a theoretical model for microbial filtration in a porous medium based on a moment balance method. The fate of an adhered microbial cell after collision with a sand collector was determined by competition between the adhesive moment due to the surface adhesion and the detachment moment due to hydrodynamic interaction. The new model takes into account the effects of flow condition, salt concentration and bacterial micro-properties. The theoretical results agree well with the
experimental results, and we believe the new model captures the fundamentals of the problem.

In summary, this dissertation shows the significance of fluid and contact mechanics in microbial/colloidal adhesion and transportation. It reveals the underlying physics and provides a valuable tool—the microfluidic test—for studying the microbial/colloidal filtration problem.
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Chapter 1. Introduction

Humans strived to clean water long before they learned to have controlled fire and had cooked food. As early as in 1500 BC, the Egyptians discovered the principle of coagulation and applied the alum for suspended particle settlement (Engineers, 1985; Viessman, Hammer, Perez, & Chadik, 2009), as shown in the Figure 1.1a. After 500 BC, Hippocrates, the Greek physician, invented the practice of sieving water and created the first bag filter (Engineers, 1985), later known as Hippocratic sleeve and schematic sketch is shown in Figure 1.1b. The major concerns of water treatment were merely the aesthetic properties of the water till late 19th Century when a cause and effect relationship between invisible microbes and disease was established thanks to advances in science and technology (Engineers, 1985; Viessman et al., 2009). In the 1890s effective water treatment techniques began to develop and America started building large sand filters. Nowadays these turns out to be a success, as shown in Figure 1.1c. Coagulation and rapid sand filtration reduced both turbidity and bacteria in water supplies significantly (Engineers, 1985).

Figure 1.1. (a). Sketch of Ancient Egyptian Water Purification device found on the wall of the tomb of Amenophis II ta Thebes, caved in 1450 B.C (Engineers, 1985). (b). Hippocratic Sleeve in 450 B.C. (c). GreenLeaf Rapid Sand Filtration System built in late 1990s in Hanoi City, Vietnam
Ironically, as developments in various aspects of human society, clean water become harder to get access to instead of easier. Now the water crises is one of the leading global risks (Forum, 2016; Unicef, 2015). According to annual report of World Health Organization in 2015, Diarrhea is the third leading cause of child death, a majority of which are water-related (Unicef, 2015). Access to safe water is crucial and urgent for both urban and rural population today, especially in developing regions. Building, operating and maintaining large infrastructure for water supplying system and water treatment plant is not time efficient and economically unrealistic in these regions. On the other hand, the pollution of water source/soil is threatening human life worldwide. Polluted pathogens can transport to safe water source through underground such as what is indicated in Figure 1.2 by polluted river bed. According to the government report in 2016, 80 percent of underground water from shallow wells in China is unsafe for drinking because of pollution from heavy metals and agricultural chemicals. In-situ bioremediation is a proven technology to reduce groundwater/subsurface soil pollution (Atlas & Philp, 2005).

In the research community, understanding transport and immobilization of contaminated agents/pathogens in granular medium is fundamental of natural/artificial filtration system and bioremediation technologies such as

Figure 1.2. Pathogens or other contaminants can penetrate the riverbank and transport in underground, potentially resulting in pollution in underground medium and groundwater. On the other hand, riverbank filtration can remove pathogens and other contaminants by porous medium of sand and improve groundwater quality in a sustainable way.

those for remediating heavy metal particles (e.g. uranium, chromium and technetium) (Tufenkji & Elimelech, 2004b). The granular medium filtration techniques, collaborating with statistical analysis, has been adopted by researchers and gained their popularity since early 1930s (Iwasaki, Slade, & Stanley, 1937). Thanks to the development in microscopy in nanotechnology and biology, e.g. Atomic Force
Microscopy (AFM), Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM), studies in microscale and nanoscale in the adsorption mechanisms were enabled. These studies revealed new factors affecting colloidal or bacterial filtration, e.g. bacterial surface characterizations (Abu-Lail & Camesano, 2003; Burks et al., 2003; Kuznar & Elimelech, 2005; Rijnaarts, Norde, Bouwer, Lyklema, & Zehnder, 1995, 1996b; Rijnaarts, Norde, Lyklema, & Zehnder, 1999; Walker, Redman, & Elimelech, 2004; Williams & Fletcher, 1996), motility (Matthew W Becker, Samantha A Collins, David W Metge, Ronald W Harvey, & Allen M Shapiro, 2004; Camesano & Logan, 1998; Camper, Hayes, Sturman, Jones, & Cunningham, 1993; Hendry, Lawrence, & Maloszewski, 1999; McCaulou, Bales, & Arnold, 1995), softness (Ivanov, 2012; Y. Li, Wang, Onnis-Hayden, Wan, & Gu, 2014; X. Wang et al., 2012), size (Matthew W Becker et al., 2004; D. Fontes, A. Mills, G. Hornberger, & J. Herman, 1991; Weiss, Mills, Hornberger, & Herman, 1995). However, these new variables increased the complexity of the problem significantly. The traditional filtration test and theoretical approaches have limitations in explaining the physics behind and integrating these new parameters into the existing models. This dissertation aims to: (i) pursue experimental approach that allows the direct investigation of coupled impact of these new factors, e.g. micro-mechanical properties of bacteria and flow rate, on filtration fate, (ii) develop a theoretical platform based on existed model to incorporate the new factors. This would guide research experiments/engineered filtration system design.

1.1 Background and Literature Review

1.1.1 Water treatment

Based on the purpose, water treatment can be divided into two types, for water supply and wastewater treatment for wastewater discharge. The first type of water treatment is used to reduce levels of contaminants in drinking water to make water safe for human consumption and/or improve the aesthetic quality of the water (Casiday, Noelken, & Frey, 2008). Untreated wastewater typically contains high levels of organic material, massive pathogenic microorganisms, as well as nutrients and toxic compounds which are environmental and health hazards. Therefore, it has to be properly treated before disposal (Tchobanoglous, 1979).

Suspended particles in public water supply usually contain: bacteria, floating debris, sand and dirt depending on the source of the freshwater. There are six major steps in the treatment of water: screening, sedimentation, precipitation, filtration,
adsorption and disinfection (Casiday et al., 2008). Two typical filters are used in filtration, gravity filter and pressure filter. Gravity filter has two types based on filtration speed, slow sand filter with flow rate of 0.1-0.4 m³/h and rapid sand filter with flow rate of 5-15m³/h (Casiday et al., 2008), as shown in Figure 1.1c. Adsorption method in treatment of public water supply also applies powdered activated carbon (PAC) and granular activated carbon (GAC). Physical, chemical and biological methods are used to remove contaminant from waste-water (Tchobanoglous, 1979). To achieve different levels of contaminant removal, individual waste-water treatment procedures are combined into a variety of systems, classified as preliminary, primary, secondary, and tertiary wastewater treatment. Natural systems are also used for the treatment of wastewater in land-based application.

Preliminary treatment prepares wastewater influent for further treatment. Screening is usually used in this step to remove gross pollutants from the wastewater and to protect downstream equipment from damage, avoid interference with plant operations. Depending on the mesh size of screen type, this method can remove objects with size from 0.001 mm to 6 mm, including large solids and rags, abrasive grit, odors, and in certain cases, unacceptably high peak hydraulic or organic loadings.

Primary treatment involves the partial removal of suspended solids and organic matter from the wastewater by means of physical operation such as screening and sedimentation. Pre-aeration or mechanical flocculation with chemical additions can be used to enhance primary treatment. Eventually, this step produces a liquid effluent suitable for downstream biological treatment and separating out solids as a sludge that can be conveniently and economically treated before ultimate disposal.

Secondary method is to remove soluble and colloidal organics and suspended solids that have escaped the primary treatment. This is typically done through biological process, namely treatment by active sludge, fixed film reactors, or lagoon systems and sedimentation. Trickling filtration is involved in this step to remove nutrition, e.g. phosphorus and Nitrogen.

Tertiary/advanced wastewater treatment is to remove significant amounts of nitrogen, phosphorus, heavy metals, biodegradable organics, bacteria and viruses. Methods include chemical coagulation, flocculation and sedimentation, followed by granular filtration and active carbon.

1.1.2 Filtration in water treatment
Many types of filtration methods are adopted in both types of water treatment. They include: screening, membrane filtration, trickling filtration, granular medium filtration (sand filtration) and activated carbon. Screening uses meshes with different sizes. Screening is usually used in the beginning of water treatment to remove large object, e.g. grit, rags or other trash. Membrane filtration is based on reverse osmosis, also named with micro filtration or ultrafiltration. It is particularly for removal of salt and other effluent material from water and usually expensive. Trickling filtration is a biological operation. The filter medium usually consists of rock or plastic packing material. The medium is coated with organisms which will consume the organic material from the wastewater when it goes through the medium. It removes organic material, e.g. Phosphorus and Nitrogen compounds. Activated carbon is a chemical adsorption. Adsorption takes place because the hydrophobic carbon surface attraction non-polar contaminant particles. This method is popular for home water purification equipment and public water supply. A noticeable limit of this method is that activated carbon is difficult to be cleaned. The activated carbon medium has to be replaced once it is saturated.

Granular medium filtration/sand filtration is applied in both large scale/natural system and lab scale for research purpose. Straining and clogging take place in this process depending on porosity and ratio of pathogen size to grain size (Matthew W Becker et al., 2004; D. Fontes et al., 1991; D. E. Fontes, A. Mills, G. Hornberger, & J. Herman, 1991; Hornberger, Mills, & Herman, 1992; M. J. Martin, Logan, Johnson, Jewett, & Arnold, 1996; Silliman, Dunlap, Fletcher, & Schneegurt, 2001; Weiss et al., 1995). A more significant mechanism is adsorption (Casiday et al., 2008) via surface interaction between pathogens and grains. This method is applied at the end of public water supply treatment and wastewater treatment. It is also applied in most natural treatment systems, like river beds. This method is also broadly applied in research community to study fate of particle/bacterial transport and adsorption in underground or soil. In this dissertation, we will focus on the granular medium filtration. The terms, porous medium filtration, granular medium filtration and sand filtration refers to the same type of experiment.

1.1.3 Porous medium filtration for research purpose
1.1.3.1 Lab Scale Test

In the lab scale, filtration test also refers to column test or leaching test. Generally, the granular medium is packed in a cylindrical column or tank which has an inlet connecting to solution to be tested and an outlet leading to a waste container. The solution is injected from inlet and flow through the column. In this process, portion of particle or bacteria would be filtered by the system. The rest would run through the system from the outlet. The particle/bacterial concentration at outlet are measured as function of time which is named as breakthrough curve (Hendry et al., 1999; Y. Li et al., 2014; Tufenkji & Elimelech, 2004a, 2004b; Tufenkji, Miller, Ryan, Harvey, & Elimelech, 2004). Depending on the type of flow, column test can be divided into flow through column test and static column test. Based on flow direction, flow through test has 3 types: up flow, down flow and inclined flow. Carefully packing medium is important in this type of test to exclude air bubbles and maintaining saturated conditions of grains. Static column test is flow free. This test method is well suited for studies on microbial attachment, taxis, chemotaxis and survival behavior. Tank experiment are typically constructed on a scale of 1 to 10 inches in length and are commonly used to study the potential of physicochemical, biological remediation of xenobiotic compounds and simulate the effects of physical or geochemical heterogeneity on subsurface bacterial transport behavior.

Compared to field scale test, column test provides a better degree of control and thus, allows a detailed investigation of specific factors affecting microbial/particle filtration, for example, flow rate, grain size and column length. However, it cannot account for coupled effects of different factors, especially more factors were reveal and proven to affect filtration. For instance, there are many factors, in addition to bacterial surface properties, which governs bacterial immobilization on grain surface. Like bacterial size, hydrodynamic interaction, bacterial softness and motility, these factors are interrelated and poor handled by column test. Another drawback is that flow velocities used in column tests are usually much higher than those commonly observed in the field (Harvey, Harms, & Landkamer, 2002). In our study, we focused on up flow through column test. A typical experimental set up and test result are shown in the Figure 1.3. Figure 1.3 (a) shows a real column test set up in our lab using a 60 cc syringe. Figure 1.3 (b) indicates a schematic of column test and Figure 1.3 (c) shows typical column test results for bacterial strain of Aeromonas punctate (strain Q), known as breakthrough curve. The bacterial concentration is measured at the column outlet as a
function of injected volume. At the initial stage, bacterial concentration at column outlet is zero, indicating bacterial trapping in the column. Bacteria started to flow through and escape the column when injected volume $\approx 1$ pv. When injected volume $\approx 2$ pv, bacterial solution is switched to background solution thus the bacterial concentration at column outlet decreases and falls back to zero as time goes to infinity.
Column packed with sands

Outlet

Flow Direction

Inlet

Spectro Photometer

Bacterial suspension

Background Solution

Waste

(b)
Figure 1.3. (a). Up flow through saturated sand packed column test. (b). Schematic of up flow through column test in lab scale. Bacterial solution is flush through the column packed with granular collectors at constant flow rate. Bacterial concentration at the outlet of column is monitored using spectrophotometer. (c). Column test results of bacterial strain of Aeromonas punctate (strain Q) under varying flow velocities.
1.1.3.2 Field Scale Test

Nature systems are highly complex environments with inherent physical, chemical and biological heterogeneity to mimic. Field scale test allows researchers to study these heterogeneities using the facilities from the field (Dong et al., 2002; Harvey, Mayberry, Kinner, Metge, & Novarino, 2002; P. Johnson et al., 2001; Schijven, Medema, Vogelaar, & Hassanizadeh, 2000; Woessner, Ball, DeBorde, & Troy, 2001). For example, several field scale studies have been used to assess the influence of chemical heterogeneities of grain (collector) surfaces (Pieper et al., 1997; Ryan, Elimelech, Ard, Harvey, & Johnson, 1999; Schijven, Hoogenboezem, Hassanizadeh, & Peters, 1999; Schijven et al., 2000). The contaminant samples are usually injected at the injection site from filed laboratory. The measurement method could be different. For some facilities, like the US Geological Survey’s Cape Cod Site, in Massachusetts (Harvey, Kinner, MacDonald, Metge, & Bunn, 1993), the monitor well at a certain distance from injection site, is applied from where the suspend (fluid-phase) microbe concentration is measured. For some facilities, like the Narrow Channel area of the South Oyster Site, in Virginia, intact cores from subsurface aquifer, at a certain distance away from injection spot, are collected. The cores are subsequently transported to the laboratory to measure the bacterial adhesion properties. The field tests are usually concentrated on particular cases of certain conditions and the results of the field studies are often difficult to generalize. Therefore, a typical usefulness of field scale test is to provide a framework in which the applicability of laboratory-derived results can be evaluated under natural conditions.

1.1.4 Factors Affecting Filtration

Due to the complexity of environmental conditions and properties of particle/pathogens, numerous factors affect the filtration process. Many of these factors are interrelated and have coupled effects. This further increases the complexity of the problem. In this section, we will classify the factors into two types: environmental conditions and particle/pathogen properties.

1.1.4.1 Environmental Factors

Ionic strength in aqueous phase affect filtration via affecting colloidal/bacterial aggregation and surface charge density (Elimelech & O'Melia, 1990; Hogg, Healy, & Fuerstenau, 1966; Tufenkji & Elimelech, 2004a, 2004b). This effect was described by

Grain size and shape contribute potentially to physical straining and clogging (Tufenkji, 2007; Tufenkji et al., 2004). Bradford investigated the impact of grain size and particle size on particle transport using column packed with glass spheres (Scott A Bradford, Bettahar, Simunek, & Van Genuchten, 2004; Scott A Bradford, Simunek, Bettahar, van Genuchten, & Yates, 2003; Scott A Bradford, Yates, Bettahar, & Simunek, 2002). The results suggested that particles were removed by both surface adsorption and physical clogging for system of intermediate grain size. Bacterial transport is significantly affected by shape of grains, oblong vs. spherical (Brown, Stencel, & Jaffé, 2002). The smallest dimension of the oblong grains dominated bacterial transport.

Flow velocity is usually observed to be negatively correlated to filtration efficiency (Bergendahl & Grasso, 2000; Shen, Li, Huang, & Jin, 2007; Tong & Johnson, 2006). However when particles were deposited in strong mode the impact of flow velocity is limited (Shen et al., 2007). John’s experimental results indicated that higher velocity resulted in a higher percentage of bacteria passing through the column as well as a marked increase in number of bacteria trapped by the column (Gannon, Tan, Baveye, & Alexander, 1991). For bacteria with motility, low flow velocity has limited impacts (Grolimund et al., 1998). Fluid velocity also affects bacteria collision with collector via the blocking effect (Rijnaarts, Norde, Bouwer, Lyklema, & Zehnder, 1996a; Rijnaarts et al., 1996b). Under low flow rate, a bacterium has smaller hydrodynamic shadow and diffuses faster. Upon approaching the collector surface, bacterium is in steeper trajectories and is less affected by attached cells.

Temperature and PH affect usually not directly, but via affecting microbe inactivation in natural environments (Tufenkji, 2007). However, the impact of these
factors on rate of inactivation is not clear (Yates, Yates, & Gerba, 1988). Temperature is the most important factor that influences virus inactivation (Schijven & Hassanizadeh, 2000; Yates et al., 1988). Temperature can also affect bacterial transport by controlling its motility (McCaulou et al., 1995).

The other factors also include porosity, packing pattern, nutrition and column dimension. We will not further review these factors because these conditions were well controlled in this study and they are not our interests at this stage.

1.1.4.2 Particle/pathogens properties

The influence of cell surface biomolecules on cell transport and immobilization which includes protein, lipopolysaccharide (LPS) and extracellular polysaccharides is significant. Recent studies which attempted to improve our understanding on the role of such macromolecules in microbial adhesion, showed that the presence of surface biomolecules can either enhance (Abu-Lail & Camesano, 2003; Burks et al., 2003; Rijnaarts et al., 1995, 1996b; Rijnaarts et al., 1999; Walker et al., 2004) or hinder (Burks et al., 2003; Kuznar & Elimelech, 2005; Rijnaarts et al., 1995, 1996b; Rijnaarts et al., 1999; Williams & Fletcher, 1996) microbe adhesion in aqueous media. Correlation of transport/adhesion behavior of E Coli (K12 mutant JM109, Abu-Lail and Camesano) and LPS on cell wall is still not clear (Burks et al., 2003; Walker et al., 2004). Proteins on the C. parvum oocyst wall can give rise to a steric interaction (repulsion) thereby decreasing the degree of oocyst attachment to quartz surface. Pili and flagella on bacterial surface could also enhance bacterial adhesion (Jacinta C Conrad et al., 2011; Gibiansky et al., 2010). However, these cell hairs can also increase the separation between cell and collector surface which could result in weakening of the van de Waals forces. The resultant effect is still not clear.

Regarding microbial filtration study, microbial growth stage and inactivation were also investigated (Y. Li et al., 2014; X. Wang et al., 2012). As mentioned in the previous section, these factors are inter-related to other factors, e.g. temperature, nutrition and PH. In some models the process of microbial growth and inactivation are described as source and sink terms respectively (Harvey, Mayberry, et al., 2002). It is difficult to find a general mechanism to describe effect of microbial growth. It should be treated on a case-by-case basis.

Particle/bacterial size and shape also affect filtration efficiency. Physical straining and clogging could play a significant role when the ratio of the particle
diameter to the medium grain diameter is greater than 0.05. Some studies showed that this critical ratio could be as low as 0.002 (Scott A Bradford et al., 2004; Scott A Bradford et al., 2002; Tufenkji et al., 2004). Note that Tufenkji (Tufenkji et al., 2004) tested the contribution of physical straining in removal by using DI water. With grain diameter of 0.21 mm, degree of removal for particle with diameter from 0.32-1.9 μm is minor. However, the 4.1 μm particle exhibits significant removal which indicates that straining can be an important capture mechanism in this situation. Weiss et al., (Weiss et al., 1995) observed preferential removal of long, rod-shaped cells. They did not consider straining as an important removal mechanism in their experiments because of the small diameter ratio between bacteria and grain (0.0014). Bradford et al. (Scott A Bradford et al., 2003) added straining on particle straining in their governing equation.

Detachment or release after trapping could be overlooked in some studies. An evidence of such release is the observation of “tailing” in measured breakthrough curves (Y. Li et al., 2014; Redman, Walker, & Elimelech, 2004; Tufenkji & Elimelech, 2004a; Tufenkji et al., 2004). A long tail is indicative of slow release. Several physical, chemical and biological factors may influence the rate and extent of microbial detachment, like bacterial motility (Matthew W Becker et al., 2004; Camesano & Logan, 1998; Camper et al., 1993; Jacinta C Conrad et al., 2011; Gibiansky et al., 2010; Hendry et al., 1999; McCaulou et al., 1995), bacterial size and Young’s modulus. Some studies indicated that detachment may be linked to nutrient availability (Tufenkji, 2007). However, this can only explain the detachment in long term. Microbial residence time also affects detachment. This cannot explain the detachment in a short term either. In our study, we focus on particle/bacterial physical factors, e.g. shape, size, surface adhesion and Young’s Modulus.

1.1.4.3 Correlation Between Bacterial Micro-Properties and Macro-Filtration Behaviour

With developments of microscopy in nanotechnology and biology, many properties of microbes and particles, e.g. shape, size, Young’s modulus and surface characterization were measured and proven to affect their filtration behavior. Xin et al. (Y. Li et al., 2014; X. Wang et al., 2012) concluded a strong correlation between bacterial micro properties, measured by atomic force microscopy (AFM), and its macro filtration efficiency measured by column test. The micro-macro properties were bridged by a modified Tabor’s parameter. Derivation of modified Tabor’s parameter is briefly
introduced in the following paragraph. Figure 1.4(a) shows AFM topographical scans of typical representative bacterial samples, conducted by Xin (Xin Wang, 2013). All strains possess similar prolate geometry with circular cross-section. The long and short axes are denoted by $b_1$ and $b_2$ respectively. Figure 1.4 (b) and Figure 1.4 (c) are cross-section profiles and 3-D topography of strain A.

(a)
A 1 mL suspension of bacteria culture in 3 mL KCl solution was pipetted onto a gelatin-treated, cleaved mica disk (Sigma G-6144) for AFM indentation. The mechanical response was derived by loading and unloading of the AFM tip. From the mechanical response, the following quantities were derived: \( b \) is the width of the ellipsoidal bacteria, \( l \) is the thickness of the cellular surface substances (CSS), \( E \) is the bacterial elastic modulus, \( U_{ad} \) is the total adhesion energy needed to detach the AFM tip from bacteria, \( R_{AFM} \) is the AFM tip radius and \( v \) is the Poisson’s ratio. More detailed measurement procedure could be found from Xin’s publication (Y. Li et al., 2014).

With the attempt to derive a universal dimensionless parameter to reflect the bacteria-surface interaction quantities as well as the bacterial dimension and mechanical properties, the modified Tabor parameter (X. Wang et al., 2012), \( \mu \), was defined by collectively combining the measured quantities listed above as

\[
\mu = \left[ \frac{2b}{l^3 E^2} \left( \frac{U_{ad}}{\pi R_{AFM}^2} \right)^2 \left(1-v^2 \right)^2 \right]^{1/3} \quad (1-1)
\]
<table>
<thead>
<tr>
<th>Strain</th>
<th>Short axis, ( b ) (nm)</th>
<th>Long axis, ( L ) (nm)</th>
<th>Elastic Modulus, ( E ) (kPa)</th>
<th>Thickness of CSS, ( l ) (nm)</th>
<th>Adhesion energy, ( U_{ad} ) (10(^{-18}) J)</th>
<th>Tabor’s parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. Raoultiella ornithinolytica (rod)</td>
<td>796 ± 300</td>
<td>1580 ± 390</td>
<td>153 ± 18</td>
<td>294 ± 101</td>
<td>212 ± 105</td>
<td>8.8 ± 0.8</td>
</tr>
<tr>
<td>SH1, Shewanella oneidensis (rod)</td>
<td>505 ± 70</td>
<td>1150 ± 410</td>
<td>249 ± 31</td>
<td>243 ± 55</td>
<td>536 ± 29</td>
<td>12 ± 4</td>
</tr>
<tr>
<td>Q. Aeromonas punctate (rod)</td>
<td>775 ± 380</td>
<td>1280 ± 700</td>
<td>105 ± 31</td>
<td>266 ± 52</td>
<td>281 ± 72</td>
<td>15 ± 4.6</td>
</tr>
<tr>
<td>H, Bacillus cereus (rod)</td>
<td>734 ± 110</td>
<td>2160 ± 310</td>
<td>211 ± 88</td>
<td>103 ± 45</td>
<td>964 ± 213</td>
<td>54.2 ± 2</td>
</tr>
</tbody>
</table>

Table 1.1 Bacterial micro properties determined based on AFM analysis and Tabor’s parameter. All strains were from stationary growth stage.

All measured quantities and modified Tabor’s parameter \( \mu \) of four bacterial strains are listed in the Table 1.1. A more detailed description of the basis and development of the modified Tabor’s Parameter can be found from our previous publications (X. Wang et al., 2012). Bacteria with a larger Tabor parameter indicates it is larger, more compliant and stickier. Therefore, once bacteria with large \( \mu \) adhered on a surface we expect it is more difficult to be detached due to its’ compliant profile, large contact area with surface and strong adhesion. This hypothesis was supported by the column test which measure the filtration efficiency of individual bacteria. The results shown a significant correlation between Tabor parameter of specific bacterial species with its’ filtration efficiency (Y. Li et al., 2014), shown in the Figure 1.5. The four strains investigated in this study are ranked as A< SH1< Q< H in terms of Tabor Parameter.

In this study, we examined the correlation of modified Tabor’s parameter and bacterial filtration efficiency measured by microfluidic test which will be discussed in the next section. We further developed the modified Tabor’s parameter by including the interaction introduced by flow velocity.
1.1.5 Filtration Theory

Since filtration method has been developed for water treatment, evaluating filtration efficiency, studying contributions of different factors involved and understanding the physics behind were always the goals of researchers. In this section, we will review the filtration theories that has been developed along with porous medium filtration test.

In natural and engineered aqueous systems, the fate of colloidal/bacterial movement, arrestment and release is believed to be controlled by the following mechanisms independently and simultaneously: (i) transport, (ii) exchange between the liquid phase and solid phase (due to attachment and detachment). Most of research has been conducted were aimed to tackled these two issues. Iwasaki conducted handful tests on sand filtration to study density distribution of pathogen in the system (Iwasaki et al., 1937). In this study, Iwasaki utilized an appropriate mathematical description for pathogen transport. By setting an absorption coefficient, the efficiency of filtration system was calibrated by filtration experiment. Though the trapping mechanism was not clear. This method was widely adopted by later researchers to study filtration efficiency of a filter system (Elimelech, 1994; Hendry et al., 1999; Rajagopalan & Tien, 1976; Tufenkji & Elimelech, 2004a, 2004b; Yao, Habibian, & O'Melia, 1971).

![Diagram of Filtration Efficiency vs. Tabor's Parameter]

- **Des**: Desulfovibrio vulgaris
- **H**: Bacillus cereus
- **SH1**: S. oneidensis MR-1
- **SH2**: S. putrefaciens CN32
- **A**: Raoultella ornithinolytica
- **Q**: Aeromonas punctata

Growth stage

\[ R^2 = 0.822 \]
\[ \frac{C}{C_0} = 0.708 \log_{10}(\mu) - 0.223 \]

100% filtration

0% filtration
Figure 1.5. Correlation between Tabor’s parameter, $\mu$, based on AFM measurement and filtration efficiency, $\alpha$, from up flow through column test (Y. Li et al., 2014).

Fundamental formula to describe transport and fate of microorganisms in porous medium is expressed as (Iwasaki et al., 1937; Rajagopalan & Tien, 1976; Schijven & Hassanizadeh, 2000; Tufenkji & Elimelech, 2004b; Yao et al., 1971; Yates et al., 1988):

$$\frac{\partial C}{\partial t} + \frac{\rho_b}{\varepsilon} \frac{\partial S}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \quad (1-2)$$

where $C$ is the concentration of aqueous phase microorganisms at a depth of $x$ and time $t$, $D$ is hydrodynamic dispersion coefficient, $v$ is initial pore velocity, $S$ is microbe concentration trapped on sand, $\rho_b$ is the dry bulk density of the porous medium, and $\varepsilon$ is the bed porosity. This equation describes, in a filtration bed, the suspended solution concentration varies as a function of time in depth $x$ of the filtration bed. The concentration distribution in radial direction of column was assumed to be uniform due to high aspect ratio of column. The first term on the right side of the equation represents the concentration change due to diffusion and the second term represents the concentration change due to convection. The first term on the left side of equation describes instantaneous concentration change at depth of $x$. The second term describes the number of suspended matters that are arrested by the collector in the controlled volume. Therefore, if we replace the second term with a adsorption/trapping term, by setting an adsorption/trapping coefficient (some study (Hendry et al., 1999) also take into account re-detachment, thus they have an extra release/detachment coefficient), we can derive this coefficient by fitting it with experimental results.

Researchers have been defining their own trapping/adsorption coefficients based on their individual experiment setup. Although these coefficients are slightly different in definition and physical meaning, they all indicate filtration efficiency of a system or colloid/pathogen. These coefficients includes: (i) $c$: coefficient of impediment modulus (Iwasaki et al., 1937), (ii) $\alpha$: attachment efficient (Shen et al., 2007; Tufenkji & Elimelech, 2004a, 2004b; Tufenkji et al., 2004; Yao et al., 1971). Attachment efficiency is defined as ratio of colloidal number that a single collector successfully trapped to the colloidal number that a collector collided with. (iii) $k_{att}$: forward, attachment, deposit coefficient. $k_{det}$: reverse, detachment, release coefficient.
Usually forward and reverse process occurred in a system simultaneously. The overall filtration efficiency is determined by competition between the two. When forward coefficient overcomes reverse coefficient, the adsorption dominates the filtration process. In this study, we will adopt attachment efficient.

The theoretical studies discussed above can be categorized into two types: (i) numerically/analytically solving the advection dispersion Eq(1-2), to study the transport of colloids/pathogen in porous medium (Elimelech, 1994; Hendry et al., 1999; Rajagopalan & Tien, 1976) and number of particles strike the collector surface (Tufenkji & Elimelech, 2004a; Yao et al., 1971), (ii) presented a physical model to interpret trapping/releasing mechanism and substitute it into the trapping terms in the Eq(1-2). In the following sections, we will present some classical models involved in the two types of studies.

1.1.5.1 Kinetic attachment and detachment processes

This is a type (ii) study. The trapping/adsorption process is assumed to be controlled by the attachment/forward and detachment/reverse rate. Therefore:

\[
\frac{\partial b}{\partial t} = k_{att} C - \frac{\rho_b}{\varepsilon} k_{det} S
\]  

(1-3)

where \(k_{att}\) and \(k_{det}\) are attachment and detachment rate coefficients respectively. This perspective is widely adopted by chemistry community, taking adsorption process as a chemical reaction. When attachment/forward rate coefficient is higher than detachment/reverse rate coefficient, the process is dominated by adsorption. The parameters, \(k_{att}\) and \(k_{det}\) are determined by experiments (Hendry et al., 1999; Iwasaki et al., 1937; Tufenkji & Elimelech, 2004a, 2004b).

1.1.5.2 The irreversible attachment under “clean-bed” conditions

This is mainly a type (ii) study. But it also collaborated with other model that involved type (i) study and came up with a complete model to evaluate filtration efficiency, for example the classical colloidal filtration theory (CFT). Here we only focus on this method and we will discuss the CFT in the next section. This model is a special case of kinetic attachment/detachment model introduced in the previous section, since the detachment process is neglected in this model. In the other words, the attachment of colloid/microbe to the collector surface is irreversible. Therefore, the Eq(1-3) was written as:
\[
\frac{P_b}{\varepsilon} \frac{\partial S}{\partial t} = k_{\text{att}} C
\]  
(1-4)

Let us make further assumptions: (i) the system may be considered at steady-state (ii) system is initially free of microorganisms (“clean-bed” condition) (iii) dispersion term is small compared to the advection term. Thus, Eq(1-2) can be solved by substituting Eq(1-4) into it as:

\[
C(x) = C_0 \exp\left[ -\frac{k_{\text{att}}}{v} x \right]
\]  
(1-5)

1.1.5.3 The Classical CFT (Colloidal Filtration Theory)

This is a type (i) study and present in 1971 by Yao (Yao et al., 1971). Yao stated three main mechanisms that affect transport of colloids from bulk solution to the collector surface, diffusion, interception and gravity. Thus, the ratio of colloid transported to a single collector surface determined by each individual mechanism was derived by solving Eq(1-2) analytically as:

\[
\eta_D = 0.9\left( \frac{kT}{\pi d_p d_c v_0} \right)^{2/3}
\]

\[
\eta_I = \frac{3}{2} \left( \frac{d_p}{d_c} \right)^2
\]  
(1-6 a-c)

\[
\eta_G = \frac{(\rho_p - \rho) g d_p^2}{18 \eta v_0}
\]

Equations (1-6 a-c) indicate contact efficiency due to dispersion, interception and gravity respectively where \( k \) is the Boltzmann constant, \( T \) is the absolute temperature, \( \tau \) is the dynamic viscosity of the fluid, \( d_p \) is the diameter of the particle, \( d_c \) is the diameter of media grains, \( v_0 \) is the flow velocity, \( \rho \) is the density of the fluid and \( \rho_p \) is the density of the particle. \( \eta_D \), \( \eta_I \) and \( \eta_G \) are single collector efficiency due to the diffusion, interception and gravity respectively. The summation of Equations (1-6 a-c) was defined as the single collector contact efficiency:

\[
\eta = \eta_D + \eta_I + \eta_G
\]  
(1-7)

This represents the ratio between number of particles that strike the collector and number of particles that flow toward the collector.
1.1.5.4 Improved CFT

This work is a type (i) study. Elimelech resolved Eq(1-2) in 1994 taking into account the van der Waals force and electrostatic double layer interaction (Elimelech, 1994). These two external forces were added into the adsorption term in Eq(1-2). The boundary condition at collector surface was considered as perfect sink. Thus Eq(1-2) was solved numerically. Using the numerical results derived in Elimelech’s work, in 2004, Tufenkji and Elimelech (Tufenkji & Elimelech, 2004a) developed a new correlation equation similar with Eq(1-6 a-c) but included vdW forces and hydrodynamic interaction as

\[
\eta_D = 2.4A_s^{1/3} N_R^{-0.083} N_{Pe}^{-0.715} N_{vdW}^{0.052}
\]

\[
\eta_I = 0.55A_s^{1/3} N_R^{1.675} N_A^{0.125}
\]

\[
\eta_G = 0.22N_R^{-0.24} N_{vdW}^{0.053} N_{gr}^{1.11}
\]

Equations (1-8 a-c) indicate contact efficiency due to dispersion, interception and gravity respectively. The dimensionless parameters in Equations (1-8 a-c) are defined in the Table 1.2.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition</th>
<th>Physical interpretation</th>
</tr>
</thead>
<tbody>
<tr>
<td>(N_R)</td>
<td>(d_p/d_c)</td>
<td>aspect ratio</td>
</tr>
<tr>
<td>(N_{Pe})</td>
<td>(vd/D)</td>
<td>Peclet number characterizing ratio of convective transport to diffusive transport</td>
</tr>
<tr>
<td>(N_{vdW})</td>
<td>(A/kT)</td>
<td>van der Waals number characterizing the ratio of van der Waals interaction energy to the particle’s thermal energy</td>
</tr>
<tr>
<td>(N_{gr})</td>
<td>(4 \frac{\pi a_p^4 (\rho_p - \rho)g}{3 kT})</td>
<td>gravitational number; ratio of particle’s gravitational potential when located one particle radius from collector to particle’s thermal energy</td>
</tr>
<tr>
<td>(N_A)</td>
<td>(\frac{A}{12\pi a_p^2 \nu})</td>
<td>attraction number; represents combined influence of van der Waals attraction forces and fluid velocity on particle deposition rate due to interception</td>
</tr>
<tr>
<td>(N_G)</td>
<td>(\frac{2 a_p^2 (\rho_p - \rho)g}{9 \pi \nu})</td>
<td>gravity number; ratio of Stokes particle settling velocity to approach velocity of the fluid</td>
</tr>
</tbody>
</table>

Table 1.2 Dimensionless parameters governing particle collision

Here \(D\) is the duffusion coefficient, \(a_p\) is the radius of the particle and \(A\) is the Hamaker constant. Note that in the new correlation equations, vdW and hydrodynamic interaction were taken into account in terms of \(N_{vdW}\) and \(N_{Pe}\). A summation of equations (1-8 a-c) was defined as single collector contact efficiency:
\[ \eta = \eta_D + \eta_I + \eta_G \]  

(1-9)

1.1.5.5 “Clean-Bed” Filtration Models with Attachment Efficiency

This is a type (ii) study. It mainly focuses on how to embed attachment efficiency \( \alpha \) into trapping term in Eq(1-2). For \( k_{att} \) and \( k_{det} \), this is straightforward as described in the section 1.1.5.1 and 1.1.5.2. For \( \alpha \), this is a bit tricky. The assumptions in Section 1.1.5.2 are still required in this method, an extra assumption is that the system is packed with uniform sized, spherical collectors. Therefore, in a unit volume of system, the number of colloids/microbes trapped can be written as (Logan, Jewett, Arnold, Bouwer, & O'Melia, 1995; Yao et al., 1971):

\[
\frac{P_b}{\epsilon} \frac{\partial S}{\partial t} = \frac{(1 - \epsilon)}{\pi/6 d_c^3} \frac{\pi}{4} d_c^2 C \eta \alpha
\]  

(1-10)

Here the term at left hand side of equation is the trapping term in Eq(1-2), \( d_c \) is collector diameter, \( \eta \) is number of colloid strike a single collector and \( \nu \) is pore velocity. Substitute Eq(1-10) into Eq(1-2), attachment efficiency can be derived as (Tufenkji & Elimelech, 2004a, 2004b; Yao et al., 1971):

\[
\ln\left(\frac{C}{C_0}\right) = -\frac{3(1 - \epsilon)}{2d_c} \eta \alpha
\]  

(1-11)

Here \( C/C_0 \) is derived from experiment and \( \eta \) is calculated as in section 1.1.5.4. Note that the inter-relationship between \( k_{att} \) and \( \alpha \) is obtained as:

\[
k_{att} = \frac{3(1 - \epsilon) \nu}{2d_c} \eta \alpha
\]  

(1-12)

1.1.5.6 Dual Deposit Mode Model

This is a type (ii) study. The CFT breaks down due to surface charge heterogeneities (Tufenkji & Elimelech, 2004b). It indicates a distribution in attachment rate. Three types of deposition may occur: (i) Particles may overcome a repulsive energy barrier to deposit in the primary energy well, (ii) Particle may deposit in a deep secondary energy well and (iii) Particles may deposit in the primary minimum in the absence of repulsive energy barrier due to surface charge heterogeneities.

1.2 Problem Statement and Research Objectives

Literally tens of thousands of contamination agents, e.g. microorganisms and particle/colloids, cause dire, global environmental and health problems. In addition, the
environmental conditions such as temperature, groundwater flow rate and fluid properties vary from region to region. Column test usually performs poorly in delineation of individual factors, especially for microorganism properties like motility, Young’s modulus, shape, surface properties and size, let alone the interrelationship of these factors. Consequently, it is extremely time consuming, expensive and literally impractical to scrutinize individual pathogens/colloids and environmental conditions using traditional column test and filtration theories.

A recent atomic force microscopic (AFM) measurement indicated that microscopic characterization of single bacterial cell and environmental conditions (ionic concentration of groundwater) are strongly correlated to bacterial macroscopic transportation/attachment behaviour by addressing the fundamental science and mechanics underlying these processes (Y. Li et al., 2014; X. Wang et al., 2012). We are motivated to develop a new technique to characterize bacterial/particle filtration behaviour more time efficiently and economically, and to provide reliable bioremediation design guidelines for practical filtration parameters. Thus in this project we plan to: (i) develop a new experimental method to overcome the shortcomings of conventional column test, (ii) construct a new dimensionless parameter account for coupled impacts of bacterial properties and flow conditions, (iii) construct a new theoretical model predicting filtration efficiency account for bacterial microcharacterizations and environmental conditions, (iv) validate the new model through macro-scale experiments with variety bacterial strains under varying ionic concentrations and flow velocities.

1.3 Organization of the Dissertation

This dissertation is organized in eight chapters.

Chapter 1 is an introduction to filtration experiment and theory in water treatment industry and research field. Relevant publications were reviewed in particle/pathogens transport and filtration in porous medium. The limitations of traditional approaches were discussed. Project objectives were proposed, motivated by the previous research findings.

In Chapter 2, we introduce the classical contact models and DLVO theory because these will be the foundation we will build our model on. A detailed derivation of contact model between particle/bacterial and flat surface in the presence of electrolyte is presented based on the classical DLVO theory and Derjaguin-Muller-
Toporov (DMT) contact model (B. V. Derjaguin, Muller, & Toporov, 1975; Muller, Derjaguin, & Toporov, 1983). Three methods are adopted and compared. With experimental results, we will discuss which method is closer to the reality.

In Chapter 3, we present a theoretical study on particle adhesion onto flat surface in humid environment. Solid-solid interaction is not considered in this study and the only inter-surface force between two surfaces is capillary force due to Laplace pressure. When relative humidity drops below 30%, meniscus height shrinks to the size of water molecules and Laplace pressure does not exist. Both classical adhesion models, the DMT model and classical Johnson-Kendall-Roberts (JKR) model (K. Johnson, K. Kendall, & A. Roberts, 1971) break down. At relative humidity of 30%-100%, a contract model for a solid sphere adhering onto a rigid substrate in the presence of long range capillary force was presented based on a force balance. The adhesion-detachment mechanics is constructed based on Hertz contact theory and Laplace-Kelvin equation. “Pull-off” is predicted with the critical tensile force larger than the DMT limit that serves as only in saturated environments.

We conducted handful flow-through packed sand column test under varying flow rate, ionic strengths and bacterial species. In Chapter 4, we presented the test results and investigated the coupled effect of flow rate and ionic strength on bacterial filtration. Three different types of bacterium were studies, Aeromonas punctate (strain Q), Bacillus cereus (strain H) and Raoultella ornithinolytica (strain A). The range of experimental parameters in this study covered the previous test parameters and thus we compared the two for validation purpose. The correlation between $\mu$ and $\alpha$ was further studied under higher flow rate. A theoretical analysis based on moment balance between fluid drag and adhesion was carried out and showed a great potential of interpreting how flow velocity affects bacterial detachment under varying Derjaguin, Landau and Verwey, Overbeek (DLVO) interaction.

Chapter 5 introduces a new experimental method on particle filtration and surface attachment using microfluidics to overcome the limits of conventional column test method. The microfluidic device allows greater control of environmental conditions and differentiates surface adhesion from other filtration mechanisms, like clogging and physical straining. Varying flow rates and ionic concentrations are studied. Coupled effects of flow rate and ionic concentration on particle attachment-detachment are found. Two deposit modes, strong deposit mode (1min) and weak deposit mode (2min), were proven to exist. Fraction of particle in the two modes under varying ionic
concentration of 3 to 100 mM KCl (aq) were experimentally derived. Filtration efficiency was directly measured. A critical flow rate was found beyond which effect of flow rate on particle detachment is limited. A theoretical study was conducted using the contact model we derived in Chapter 2. The calculated results are consistent with experimental findings. Magnitude of adhesion in the 1 min is significantly higher than adhesion in the 2 min and detachment due to the hydrodynamic drag. Magnitude of adhesion in 2 min is comparable to magnitude of detachment due to hydrodynamic drag.

In Chapter 6, we extended the microfluidic study introduced in Chapter 5 to four different bacterial strains under a broad range of flow velocity, $1.5\times10^{-6}$ m/s to $1.5\times10^{-3}$ m/s and constant ionic concentration of 3 mM KCl (aq). Microfluidic test results showed a good agreement with the classical column test results. The strong correlation between physical properties of bacteria and its attachment efficiency were derived using the microfluidic device. Hydrodynamic interaction has a clear impact on bacterial detachment except for one. A universal predictor of bacterial filtration efficiency, addressing the coupled impact of flow velocity and bacterial micro-properties, was developed based on the modified Tabor’s parameter (Y. Li et al., 2014; X. Wang et al., 2012). The new predictor revealed the interrelationship between environmental conditions and bacterial micro-properties and potentially provide guidelines for water treatment and research study on bacterial attachment-detachment in the presence of fluid.

Chapter 7 introduces a new theoretical model on bacterial filtration through porous medium. For clean bed filtration, surface adhesion dominates the adsorption mechanisms. The filtration process is thought to have two steps: bacterial transport in aqueous phase and bacterial adsorption in solid phase (Tufenkji, 2007). Bacterial transport around a single spherical collector was described by the convection-diffusion equation (1-2). The collision probability of bacteria on different regions on collector surface was derived by solving the governing equation. Therefore, the moment balance method used in the Chapter 5 was adopted here to determine attachment-detachment of a single bacteria. We developed a cylinder contact model in the presence of the DLVO interaction which gives the adhesive torque applied on attached bacteria. The hydrodynamic torques of varying inclination angle were derived using COMSOL. Micro-properties of six bacterial strains and environmental conditions, e.g. flow velocity and ionic concentration, were used as input parameters in the model. The results of new model were compared with the column test results. The new model
revealed the physics behind the bacterial adsorption and provides a theoretical scheme allowing investigation of multiple factors, e.g. biological factors, environmental factors and mechanical properties and their interrelationship.

Chapter 8 summarized the significant contributions and conclusions of this study. The possible directions of future works are also proposed.
Chapter 2. Adhesive Model in the Presence of Electrolyte

2.1 Introduction

In this study, we focus on surface adhesion among multiple bacterial/particle adsorption mechanisms in filtration. We assume that in dry environment the adhesive contact between two objects, particles/pathogens and collectors, originates from the interaction of individual atoms. These interatomic interactions are characterized by strong, short range and attractive van der Waals interaction. When the contact is presented in electrolyte, electrostatic interaction comes into play. Accumulation of charged ions on object surface give rise to surface electrical potential and form an electrical double layer (EDL). This phenomenon is described by the Derjaguin, Landau, Verwey and Overbeek (DLVO) theory (Elimelech & O'Melia, 1990; Gregory, 1981; Hogg et al., 1966; van Smoluchowski, 1917). Interaction between two objects due to EDL potential is characterized by weak, long range and repulsive electrostatic interactions. In this Chapter, classical adhesion models in dry environment are firstly reviewed and then followed by an introduction of DLVO theory. Three methods of calculating surface force due to DLVO are presented. At the end of this Chapter, a discussion of adhesion model in the presence of electrolyte is presented based on different type of adhesion models. Note that the adhesion mechanisms in contact between microbes and flat surface are more complex than just van der Waals interaction. For example, pilus and flagella also enhance bacterial bonding on a surface (Jacinta C Conrad et al., 2011; Gibiansky et al., 2010). However instead of differentiating individual adhesion mechanisms, the goal of this dissertation is to build a theoretical platform that takes into account the magnitude and effective range of a universal surface interaction so that other type of surface interactions could be included into the platform in the future. At this stage of study, we take van der Waals interaction as the only surface interaction.

2.2 Solid Adhesion Models

2.2.1 van der Waal Attractive interaction

van der Waals (vdW) interaction is in nature due to electromagnetic force between atoms, molecules or ions. Attractive intermolecular interactions consist of four types: dipole-dipole interaction (known as Keesom forces), ion-dipole interactions,
dipole-induced dipole interactions (known as Debye forces), and instantaneous dipole-induced dipole interactions (known as London dispersion forces). Among these, the combination of Keesom, Debye and London dispersion forces are known collectively as the van der Waals attraction (Israelachvili, 2015). In 1924, John Lennard-Jones derived a general mathematically model to approximate the interaction between a pair of neutral atoms or molecules, combining the effect of van der Waals attraction and the electron cloud overlap repulsion (J. E. Jones, 1924).

There are, essentially, two approaches to compute the van der Waals interaction. Firstly, the interaction potential between macroscopic bodies is derived by a pairwise summation of the relevant intermolecular interactions discussed above (Gregory, 1981). Therefore, applying the Lennard-Jones interaction energy between two atoms or molecules, the Lennard-Jones disjoining pressure per unit area between two objects is given by

\[
p_{LJ}(z) = \frac{8 \Delta \gamma}{3 z_0} \left( \frac{z_0}{H} \right)^3 - \left( \frac{z_0}{H} \right)^9
\]

(2-1)

Here \(z_0\) is interatomic equilibrium spacing, \(H\) is separation between two surfaces and \(\Delta \gamma\) is the surface energy of adhesion two objects which depends on material properties. Due to the nature of electromagnetic interaction, there is a finite time of propagation in the medium between objects. This results in a retardation and a reduced interaction. This effect can be significant in particle deposition at separation larger than a few nanometres. Thus second approach takes into account the retardation effect based on Lifshitz theory (Lifshitz, 1956). Despite ignorance of retardation effect, the first approach gives acceptable results for dry environment and great convenience (Gregory, 1981). Thus, in the following sections of adhesion models, surface interactions are calculated based on un-retarded vdW interaction. In the DLVO section, the retarded vdW interaction will be applied.

### 2.2.2 Hertz Contact Model (No Adhesion)

In 1881, Hertz developed a continuum model to predict the contact area for two spherical objects (H Hertz, 1881) with radii \(R_1\) and \(R_2\), shown in the Figure 2.1 (a). Based on hypothesis: (1) the ratio of contact radius to effective radius, \(a/R\) is small; (2) no friction occurs at the interface; (3) no tensile stress exists in the contact area (unilateral contact), Hertz found that the contract radius \(a\) was related to the external
load \( P \), displacement of the two spheres centres \( \delta \), the effective radius \( R \), vertical displacement \( u_z(r) \), and the elastic properties of the contact materials as,

\[
a^3 = \frac{PR}{K}
\]

(2-2)

\[
\delta = \frac{a^2}{R}
\]

(2-3)

\[
u_z(r) = \frac{a^2}{\pi R} \left\{ \sqrt{\left(\frac{r}{a}\right)^2 - 1} + \left[ 2 - \left(\frac{r}{a}\right)^2 \right] \sin^{-1} \frac{a}{r} \right\}, \quad r > a
\]

(2-4)

with the equivalent elastic modulus, \( K \), and the effective radius \( R \) of two spheres, given by,

\[
K = \frac{4}{3} \left( \frac{1-v_1^2}{E_1} + \frac{1-v_2^2}{E_2} \right)^{-1}
\]

(2-5)

\[
R = \frac{R_1 R_2}{R_1 + R_2}
\]

(2-6)
Figure 2.1. Solid adhesion models. (a) Two elastic spheres making contact under applied load $P$, the deformation profile, indicated black shaded portion, and stress distribution, indicated by red portion, predicted by (b) Hertz model, (c) JKR model, (d) DMT model. “$a$” is the contact radius.

where $E$, $v$ and $R$ are the Young’s modulus, the Poisson’s ratio and radius of sphere, respectively, and subscripts 1 and 2 denote the two objects respectively. In the absence of adhesion, the Hertz model has been shown to accurately describe the contact area and deformed profile between two solid objects (K. Johnson, 1985). The deformed profile and “parabolic” pressure distribution are shown in the Figure 2.1 (b). Hertz relation between external load and contact radius is given in the Figure 2.2. Interfacial interaction becomes significant at small scales (K. Johnson, 1985). Therefore, adhesion arising from the attractive surface interactions must be included in the contact model. Note that $u_r(r)$ is vertical displacement here, not the gap between two surfaces. To derive the gap between two deformed surfaces, $\delta$ and original profile have to be taken into account.

2.2.3 DMT Adhesion Model

To incorporate the effect of adhesion in Hertz contact, Derjaguin, Muller and Toporov assumed that the contact profile remains the same as in Hertz contact but with interfacial attractive interactions outside the contact area (B. V. Derjaguin et al., 1975; Muller et al., 1983). The DMT theory was built based on force balance, the elastic reaction force must balance both the applied load $P$ and surface adhesive force (Pashley, 1984). The interfacial attraction follows the Lennard-Jones interaction potential and was taken as part of external loads. Surface adhesion energy density $\gamma$ was derived by integral of Lennard-Jones potential from zero to infinite. Even when applied external load is removed the contact radius is nonzero due to interfacial adhesion. “Pull-off” force to reduce contact radius to zero is $F^* = -2\pi R \gamma$. The deformation profile and pressure distribution are shown in the Figure 2.1 (c). The relation between the applied load $P$ and contact radius $a$ is shown in the Figure 2.2. The mechanisms are described by the following set of equations,

$$a^3 = \frac{R}{K} \left( P + F_{vdW} \right)$$  \hspace{1cm} (2-7)
\[ \delta = \frac{a^2}{R} \]  

(2-8)

\[ u_z(r) = \frac{a^2}{2\pi R} \left\{ \left( \frac{r}{a} \right)^2 - 1 + \left[ 2 - \left( \frac{r}{a} \right)^2 \right] \sin^{-1} \frac{a}{r} \right\}, \quad r > a \]  

(2-9)

Note that the displacement \( u_z(r) \) is the same as in Hertz model and \( F_{vdW} \) represents the surface attractive force due to van der Waals. Applied load \( P \) and \( F_{vdW} \) together taken as external load that cause sphere deformation.

### 2.2.4 JKR Adhesion Model

Johnson, Kendall and Roberts presented their adhesion model in 1971, the classic JKR theory (K. Johnson et al., 1971), using a balance between potential energy of external load, stored elastic energy and surface energy. The JKR theory assumes adhesion only exists within the contact area. The deformed profile and pressure distribution are shown in the Figure 2.1 (d). Relation between the applied force and contact radius is given in the Figure 2.2. The “pseudo-parabolic” neck formed at contact edge because of stress singularity. The mechanisms are described by the following set of equations,

\[ a^3 = \frac{R}{K} \left( P + 3R\pi \gamma + \sqrt{6\pi R\gamma P + (3R\pi \gamma)^2} \right) \]  

(2-10)

\[ \delta = \frac{a^2}{6R} \left( 1 + \frac{2P}{F_1} \right) \]  

(2-11)

\[ z = \frac{a^2}{2\pi R} \left\{ \left( \frac{r}{a} \right)^2 - 1 - \left[ 2 - \left( \frac{r}{a} \right)^2 \right] \frac{4}{3} \left( \frac{a_0}{a} \right)^2 \right\} \tan^{-1} \sqrt{\frac{r^2}{a^2} - 1}, \quad r > a \]  

(2-12)

with \( F_1 = P + 3\pi R\gamma + \sqrt{6\pi R\gamma P + (3\pi R\gamma)^2} \) and \( a_0 = \frac{3}{2\sqrt{12\pi R^2 \gamma}} \). “Pull-off” force occurs at \( F^* = -\frac{3}{2\pi R\gamma} \).

### 2.2.5 Tabor’s Parameter

The inconsistency between the results predicted by DMT and JKR triggered a long dispute between the two groups. Tabor, in 1977, proposed a dimensionless parameter \( \mu = (R\gamma^2/K^2 z_0^3)^{1/3} \), where \( z_0 (\approx 1 \text{ nm}) \) is the force range of typical van der Waals interaction and \( K \) is equivalent elastic modulus defined by Equation (2-5) (Tabor, 1977). Tabor’s parameter governs the continuous transition between the two theories.
Tabor pointed out the main limits of the two theories. DMT method neglected the deformation due to the surface attraction outside the contact area and is valid only for \( \mu \ll 1 \) (hard solids, small radius and low surface adhesion energy). Whereas JKR method neglected the adhesive force outside the contact area and is valid only when \( \mu \gg 1 \) (soft, large solids with high surface adhesion energy). Maugis adopted fracture mechanics, assumed constant pressure in the Dugdale-Barenblatt-Maugis (DBM) cohesive zone of a finite range and magnitude and derived the transition from JKR limit to DMT limit (Daniel Maugis, 1992). A transition of mechanical response from JKR to DMT limits are shown in the Figure 2.2 and is govern by a dimensionless parameter \( \lambda \) where \( \lambda = 1.157 \mu \). JKR dominates when \( \lambda > 5 \) and DMT dominates when \( \lambda < 0.1 \). The “transition regime” between 0.1 and 5 is governed by two basic equations as,

\[
\frac{\lambda \bar{a}^2}{2} \left[ \sqrt{m^2 - 1} + (m^2 - 2) \cos^{-1} \left( \frac{1}{m} \right) \right] + \frac{4 \lambda^2 \bar{a}^3}{3} \left[ \sqrt{m^2 - 1} \cos^{-1} \left( \frac{1}{m} \right) - m + 1 \right] = 1
\]

(2-13)

\[
\overline{F} = \bar{a}^3 - \lambda \bar{a}^2 \left[ \sqrt{m^2 - 1} + m^2 \cos^{-1} \left( \frac{1}{m} \right) \right]
\]

(2-14)

where \( \overline{F} \) \( \bar{a} \) are dimensionless parameter of \( F \) and \( a \) given by,

\[
\overline{F} = F / \pi R \gamma \quad \text{and} \quad \bar{a} = a / \left( \pi R^2 \gamma / K \right)^{1/3}
\]

(2-15)

The parameter \( m \) represents the ratio between the contact radius \( a \) and an outer radius \( c \) of cohesive zone.
2.3 DLVO Theory

Inter-surface potential energy between particle-particle and particle substrate in ionic solvent is typically composed of electrical double layer (EDL) repulsion and van der Waals attraction. The combination of two effects was investigated by Derjaguin, Landau, Verwey and Overbeek and known as classical DLVO interaction (B. Derjaguin, 1941; Elimelech & O'Melia, 1990; Hogg et al., 1966; Verwey & Overbeek, 1955). The DLVO interaction affects many filtration processes in nature, like colloidal stabilization and bacterial/particle transport.

2.3.1 Electrical Double Layer (DEL)

Charged or uncharged particles in electrolytic solution have electric attraction and chemical preference on their surface. Opposite charges in the electrolyte carried by
counterions are absorbed onto the particle surface and form a stiff layer of intense counterions, called stern layer or stationary layer, shown in the Figure 2.3. The counterions in the stern layer are unmovable due to strong electric attraction and usually cannot neutralize the particle surface charge completely. The remaining surface charge attracts further counterions from surrounding solution towards the particle neighborhood. Density of counterions decreases with the distance from particle surface and reaches bulk density at the distance where Coulomb force and thermal motion are balanced. This is the diffusive layer or slipping layer with counterions loosely distributed, shown in the Figure 2.3. Ions in the diffusive layer are movable.

Figure 2.3. Electrical Double Layer around a negatively charged spherical particle. The stern layer refers to the layer of positively charged ions closest to the particle surface. The diffusive layer refers to a layer of movable ions beyond which coulomb force and thermal motion are balanced.

The stern layer and the diffusive layer combined are known as electrical double layer (EDL). Accumulation of charges on particle surface gives rise to a surface charge potential which results in a repulsive interaction between two identical particles in the presence of electrolyte. The total EDL potential energy density $V$ between two infinite
flat surfaces is described as (Gregory, 1981; Hogg et al., 1966) (a detailed derivation procedure of Equation (2-16) is shown in Appendix 1)

\[
V = \frac{\varepsilon K}{2} \left[ 2\psi_{01}\psi_{02} \csc h 2\kappa d - \left( \psi_{01}^2 + \psi_{02}^2 \right) \coth 2\kappa d - 1 \right]
\]

(2-16)

where \(\varepsilon\) is dielectric constant of the medium, where \(\kappa\) is the inverse Debye-Hückel length, \(d\) is surface separation and \(\psi_{01}, \psi_{02}\) are the electric potential on the two surfaces. From Equation (2-16), the total potential energy \(V\) between a sphere and an infinite flat surface can be derived as (Hogg et al., 1966; Tabor, 1977) (a detailed derivation procedure of Eq(2-17)is shown in Appendix 1)

\[
U = \pi \varepsilon \cdot R \left( \psi_{01}^2 + \psi_{02}^2 \right) \left[ \frac{2\psi_{01}\psi_{02}}{\psi_{01}^2 + \psi_{02}^2} \ln \left( \frac{1 + \exp(-\kappa H_0)}{1 - \exp(-\kappa H_0)} \right) + \ln(1 - \exp(-2\kappa H_0)) \right]
\]

(2-17)

where \(R\) is sphere radius and \(H_0\) is separation between particle and flat surface. Note that this relationship holds based on two important assumptions: (i) surface electric potential is low, less than \(25\) mV, (ii) particle size is large compared to double layer effective “thickness”.

2.3.2 DLVO Potential and DVLO Force

The retarded van der Waals interaction between a sphere and a plate of the same material is given by (Hogg et al., 1966)

\[
U_{vdw} = - \frac{AR}{6H_0(1 + 14H_0/\lambda)}
\]

(2-18)

where \(A\) is the Hamaker constant, \(\lambda\) is the characteristic wave length of the dielectric and \(H_0\) is separation of two objects. Therefore, the total DLVO potential energy may be derived as,

\[
U_{DLVO} = \frac{AR}{6H_0(1 + 14H_0/\lambda)} + \pi \varepsilon \cdot R \left( \psi_{01}^2 + \psi_{02}^2 \right) \left[ \frac{2\psi_{01}\psi_{02}}{\psi_{01}^2 + \psi_{02}^2} \ln \left( \frac{1 + \exp(-\kappa H_0)}{1 - \exp(-\kappa H_0)} \right) + \ln(1 - \exp(-2\kappa H_0)) \right]
\]

(2-19)

Derivation of DLVO force is critical in studying colloidal stabilization and detachment/attachment from each other or substrate and understanding the mechanics behind. Here, we provide three typical methods to calculating the DLVO force.
2.3.2.1 Method-1

The straight forward method of deriving DLVO force is taking derivative of total potential energy due to DLVO interaction with respective to the separation of \( H_0 \).

The DLVO force can thus be derived as,

\[
F_{DLVO}(H_0) = \frac{\partial U_{DLVO}(H_0)}{\partial H_0}
\]

\[
= \frac{R \alpha \lambda (28H_0 + \lambda)}{6H_0^2 (14H_0 + \lambda)^2} + \frac{2 \pi \varepsilon \kappa R [\psi_{01}^2 - 2 \exp(\kappa H_0) \psi_{01} \psi_{02} + \psi_{02}^2]}{-1 + \exp(2\kappa H_0)} \quad (2-20)
\]

This method is adopted in some particle detachment models (Shen et al., 2010; Torkzaban et al., 2007). In these models, the hydrodynamic interaction and the surface adhesive interaction exerted on particles are calculated and compared such that the detachment of particles is determined.

2.3.2.2 Method-2

The total potential energy density is,

\[
V_{DLVO}(H) = -\frac{A}{12\pi H^2} + \frac{\varepsilon \kappa}{2} \left[ 2\psi_{01} \psi_{02} \coth 2\kappa H - (\psi_{01}^2 + \psi_{02}^2) \left( \coth 2\kappa H - 1 \right) \right] \quad (2-21)
\]

The first term is surface energy density due to van der Waals interaction and the second term is simply EDL potential density. The exact disjoining pressure was derived by taking derivative of the total potential energy density. Then directly integrating the exact disjoining pressure with respect to entire particle surface, the DLVO force is derived as,

\[
F_{DLVO}(H_0) = \int_0^\infty \frac{\partial V_{DLVO}(H)}{\partial H} 2\pi r \cdot dr
\]

where \( H_0 \) is the separation between the sphere and substrate, \( H \) is gap between surfaces and \( r \) is radius corresponding to \( H \) as shown in the Figure 2.1 (a). \( H \) is a function of \( r \) as \( H = H_0 + R - (R^2 - r^2)^{1/2} \).

2.3.2.3 Method-3

The disjoining pressure between a spherical particle and an infinite plane is derived as,

\[
p(z) = \frac{\partial U_{DLVO}(z)}{\pi R^2 \partial z} \quad (2-23)
\]
where \( z \) is intersurface separation and \( R \) is particle radius. Figure 2.4 shows the disjoining pressure as a function of surface separation \( z \). To further simplify the form of \( p(z) \) and highlight the underlying physics, a modified form of the Dugdale-Barenblatt-Maugis (DBM) cohesive zone approximation is adopted (Daniel Maugis, 1992), where \( p(z) \) assumes a stepwise function with linear transition as shown in the Figure 2.4 and Figure 2.5. The cohesive zone between two surfaces is divided into three layers according to the profile of DLVO interaction, the attractive primary minimum (1min) with range of \( z_1 \), the repulsive region with range of \( z_r \) and the attractive secondary minimum (2min) with range of \( z_2 \). In each layer, the disjoining pressure is set to be constant \( p_i \) such that the surface energy in the layer is preserved as

\[
p_i \times z_i = \int_{z_{i-1}}^{z_{i+1}} p(z) \, dz
\]

(2-24)

Figure 2.4. Schematic of disjoining pressure \( p(z) \) due to DLVO potential, indicated by the grey portion, and DBM cohesive zone approximation, indicated by the blue dash line.

with the subscript \( i = 1 \) and 2 for 1min and 2min respectively and \( i = r \) for repulsion. The approximated disjoining pressure is expressed as
Here $z_0$ is equilibrium spacing which is not shown in Figure 2.4 and 2.5. The total DLVO force can be derived by summation of force due to the disjoining pressure in each layer as

$$F_{\text{DLVO}} = p_1 \pi(c_i^2 - a^2) - p \pi(c_i^2 - c_r^2) + p_2 \pi(c_r^2 - c_i^2)$$

where $a$ is the contact radius and $c_i$ is the projected radius for each layer as shown in Figure 2.5.

According to our calculation, results of three methods are noticeably different. For a sphere with diameter of 1~2 \(\mu\)m in point contact with a flat surface under ionic concentration of 30 mM, method-1 suggests magnitude of DLVO force is $10^{-8}$ N, method-2 suggests the magnitude of DLVO force is $10^{-10}$ N and method-3 suggests the magnitude is $10^{-11}$ N. In this thesis, we will mainly adopt method 2 and method-3.

## 2.4 Adhesive Models with DLVO Theory

There are multiple ways to integrate the DLVO theory into adhesive model depending on the type of adhesive model, DMT, JKR or Maugis model.

### 2.4.1 Based on DMT Model

![Diagram of Dugdale-Barenblatt-Maugis (DBM) cohesive zone approximation](image)

Figure 2.5 Dugdale-Barenblatt-Maugis (DBM) cohesive zone approximation between a sphere and flat surface, consisting of three layers, 1\text{st} min attraction, repulsion barrier and 2\text{nd} min attraction. “a” is contact radius. “$c_i$” is projected radius for each layer.
Since DMT model is built based on a force balance, the DLVO force can be taken as an external inter-surface force. When zero external force is applied, the DLVO force would result in a finite contact radius and balance out the Hertz force due to particle deformation. To derive the DLVO force, we adopted the method-2 we discussed in the Section 2.3.2. We will focus on this model in the Chapter 5.

2.4.2 Based on JKR Model

The JKR model uses the minimum total potential energy approach in which total potential energy of the system is calculated, including surface energy. DLVO potential can be taken as total surface energy. A detailed implementation can be found in Jiayi’s work (Shi et al., 2013).

2.4.3 Based on Maugis Model

Maugis analysis was based on fracture mechanics and required knowing the surface stress distribution in the cohesive zone. We will adopt this model in Chapter 7, studying contact between a solid cylinder and a rigid surface in the presence of electrolyte. The surface stress in the cohesive zone is approximated by the method-3 discussed in the Section 2.3.2 as \( p(z) \).

2.5 Summary

This chapter introduced the classical contact and adhesion models in dry and inter-surface interactions in aqueous environmental. We briefly discussed the extension of the application of the adhesion model to aqueous environments which will be used in calculating surface adsorption in filtration. In the Chapter 5 and 7 we will discuss in detail the spherical and cylindrical adhesion models in the presence of electrolyte using the DMT model and the Maugis analysis respectively.
Chapter 3. Adhesion of a Solid Sphere on a Rigid Planar Substrate in the Presence of Moisture

3.1 Introduction and problem statement

Adhesion of an elastic sphere onto another sphere or a rigid substrate is ubiquitous in many branches of science and technology such as colloidal particles, storage and transportation of glass powder, and micro-electromechanical systems (MEMS) especially when the typical dimension shrinks to submicron scale (K. L. Johnson, 1985; R. Jones, Pollock, Cleaver, & Hodges, 2002; Kendall, 2001). Exposure to moist air leads to meniscus formation at the contact interface and could post serious problems of operation hindrance and reliability. Figure 3.1 (a) demonstrates the formation of a capillary neck between two B-UNCD sidewalls in a humid unprotected environment (Buja, Kokorian, Sumant, & van Spengen, 2015). This process can be observed under microscope in real time. Figure 3.1 (b) shows a typical failure mechanism of compliant structures in MEMS fabrication and packaging. Under rapid gas flow, free standing microcantilever can be brought into contact with substrate and remain adhered. The adhesion mechanism shown in the Figure 3.1 (b) is due to capillary interaction. As relative humidity (RH) increases, the adhered portion from the crack tip, indicated by red line, increases (Soylemez & de Boer, 2014).
Figure 3.1. (a) Effect of capillary formation between un-cleaned MEMS sidewalls (Buja et al., 2015). (b) Cross-section schematic geometry of the microcantilever in the “S” shape. Interferograms of cantilevers 5-14 at 29% RH and after increasing exposure to humid conditions (Soylemez & de Boer, 2014).

Meticulous experimental work conducted by Christenson (Christenson, 1985, 1988), Maugis (Maugis & Gauthier-Manuel, 1994), and Xu (Xu, Liechti, & Ravi-Chandar, 2007), and theoretical framework constructed by Fogden (Fogden & White, 1990) showed the effect of moisture on adhesion. In a relatively dry environment, the inevitable meniscus shrinks to a small dimension and the resulting short-range Laplace pressure leads to the Johnson-Kendall-Roberts (JKR) limit (Fogden & White, 1990). Close to the saturated moisture limit, the meniscus grows to such a large extent that adhesion mechanics approaches the Derjaguin-Muller-Toporov (DMT) limit (Fogden & White, 1990). Wan et. al. (Wan & Lawn, 1990) investigated adhesion of two non-interacting elastic cantilevers in the presence of meniscus and found that the critical energy release rate is twice the surface tension of water as if the crack front is right at the meniscus. The intermediate range of relative humidity is accounted for by the Tabor’s parameter that accounts for the JKR to DMT transition (Maugis, 2000). In such classical work, the “pull-off” force leading to spontaneous detachment is taken to be \( F^* = -\chi \times \pi R \times 2\gamma \), where \( R \) is the sphere radius, \( \gamma \) is the surface tension of water, and \( \chi \) is a numerical constant \( \chi_{\text{DMT}} \leq \chi \leq \chi_{\text{JKR}} \) with \( \chi_{\text{JKR}} = 3/2 \) and \( \chi_{\text{DMT}} = 2 \). Therefore \( F^* \) is a monotonic increasing function of relative humidity approaching the JKR and DMT limits in the extremes. The transition from JKR to DMT is accounted for by the Tabor’s parameter. Streator(Zheng & Streator, 2003) and Polycarpou (Xue & Polycarpou, 2007, 2008) started from a long-range inter-surface attraction and derived the sphere deformation. The numerical approach interestingly showed a monotonically decreasing function of humidity though \( F^* \) approaches the DMT limit when relative humidity is higher than 70%.

In this chapter, we will build an alternative model based on the Hertz contact theory (K. L. Johnson & Johnson, 1987) and cohesive zone model (Wan & Lawn, 1992a), rather similar to the DMT model. We will first present the theory and results, leaving the discussion and comparison with the classical models to Section 3.4. In the absence of moisture, we assume null interaction between the sphere and substrate such that only the meniscus provides an inter-surface force. The total force acting on the
sphere therefore becomes the sum of the applied load and the Laplace pressure within the meniscus. The compressive stress distribution within the contact and the deformed geometry strictly follow the classical Hertz contact theory (H. Hertz, 1896, trans. English). The Dugdale-Barenblatt-Maugis (DBM) cohesive zone model is adapted where the disjoining pressure is taken to be constant over a finite range determined by the Laplace-Kelvin equation (K. L. Johnson & J. A. Greenwood, 2008; Wan & Lawn, 1990, 1992b; Wan, Smith, & Lawn, 1992). The measurable relations of applied load, contact radius, and approach distance are derived.

3.2 Theory

A compressive load, \( F \), is applied to an elastic sphere with radius \( R \), elastic modulus, \( E \), and Poisson ratio, \( \nu \), resulting in an approach distance, \( \delta \), and contact radius, \( a \). In the absence of adhesion, the classical Hertz contact theory (H. Hertz, 1896, trans. English) gives the interrelation of \((F, a, \delta)\), and the axisymmetric deformed profile \( z(r) \), namely,

\[
\delta = \frac{a^2}{R} \quad (3-1)
\]

\[
F = \frac{4}{3} \left( \frac{E}{1 - \nu^2} \right) \frac{a^3}{R} \quad (3-2)
\]

\[
z(r) = \frac{r^2}{2R} - \delta + \frac{a^2}{\pi R} \left\{ \sqrt{\left( \frac{r}{a} \right)^2 - 1} + \left[ 2 - \left( \frac{r}{a} \right)^2 \right] \sin^{-1} \frac{a}{r} \right\}, \quad r > a \quad (3-3)
\]
Figure 3.2. An elastic solid is deformed from a spherical geometry (dashed curve) into Hertz profile (solid curve). The uniform Laplace pressure bounded by the meniscus exerts traction on the sphere in addition to the applied load.

The external load is balanced by the Hertz reaction force $F_H = F$ at the sphere-substrate interface. In the presence of a general intersurface interaction potential $\phi(z)$, a disjoining pressure $p = d\phi(z)/dz$ arises at the sphere-substrate interface, and the total adhesion force acting on the sphere becomes

$$F_{ad} (a) = \int_{a}^{\infty} \frac{d\phi(z)}{dz} \times 2\pi r dr$$

(3-4)

Here $F_{ad}$ is taken as an addition to the applied load, $F$, while Equations (3-1), (3-2) and (3-3) remain valid. Total force acting on the sphere is therefore balanced by

$$F_H (a) = F + F_{ad} (a)$$

(3-5)

The DBM approximation requires the disjoining pressure to be uniform, $p^*$, with a range, $z^*$, over a cohesive zone immediately outside the contact circle (Figure 3.2), such that
\[ p(z) = \begin{cases} 
0 & \text{for } z < z_0 \text{ and } r < a \\
p^* & \text{for } z_0 \leq z \leq z^* \text{ and } a < r < a_1 \\
0 & \text{for } z > z^* \text{ and } r > a_1 
\end{cases} \]  

(3-6)

The annular cohesive zone is bounded by \( a < r \leq a_1 \) with \( z(r=a_1) = z^* \). Substituting (6) into (4), the effective adhesion force becomes

\[ F_{ad} = p^* \times \pi \left( a_1^2 - a^2 \right) \]  

(3-7)

with the interfacial adhesion energy given by \( \gamma = p^* \times (z^*-z_0) \approx p^* \times z^* \), since \( z^* \gg z_0 \). It is apparent that \( a_1 \) depends on both \( z^* \) which depends on the interface chemistry and \( z(r) \) which depends on the elastic deformation. The measurable relations, \( a(F) \) and \( a(\delta) \), can then be solved in a self-consistent manner based on Equation (3-1) to (3-8).

In brief, for a certain interface with a fixed \( \gamma \), an initial value of \( a \) is arbitrarily chosen to solve for \( \delta \) and \( F_H \) according to Equation (3-1) and (3-2). The resulting \( z(r) \) is solved numerically for \( a_1 \), which is now the new upper limit in the integral Equation (3-4). Subsequently, \( F_{ad} \) and \( F \) are determined by Equation (3-6) and (3-7).

When the sphere is exposed to a humid environment, water vapor condenses at the contact edge forming a meniscus at the cleft. The negative Laplace pressure in the meniscus now serves as the only inter-surface attraction and is uniform within the cohesive zone given by \( p^* = \gamma / r_k \), with \( \gamma \) the surface tension (or, equivalently, surface energy) of water. The force range is close to the meniscus diameter or twice the Kelvin radius (Wan & Lawn, 1990), \( r_k \), or,

\[ z^* = 2r_k = -\frac{2v_m \gamma \cos \theta}{kT \log(1/ RH)} \approx -\frac{2v_m \gamma}{kT \log(1/ RH)} \]  

(3-9)

with a contact angle \( \theta \approx 0^o \), molecular volume of water \( v_m \), Boltzmann’s constant \( k \), temperature \( T \approx 300 \) K, and relative humidity \( RH \). At the saturation limit (\( RH \approx 100\% \)), \( p^* \rightarrow 0 \) and \( z^* \rightarrow \infty \). In dry conditions with \( RH = 0\% \), \( p^* \rightarrow \infty \) and \( z^* \rightarrow 0 \). Using (7), the adhesive force is given by

\[ F_{ad} = \frac{kT \ln(1/ RH)}{2v_m} \times \pi (a_1^2 - a^2) \]  

(3-10)

The relation \( (F, a, \delta) \) can then be found numerically by substituting (8) and (9) into (1) to (7).
3.3 Results

Figure 3.3 (a) shows $a(F)$ for a range of RH for a solid sphere with elastic modulus of 1 GPa. For $RH = 5\%$, the detachment trajectory follows path ABCDO (dark curve). At A when $F = 0$, a non-zero contact radius is expected. Increasing tensile load ($F < 0$) reduces the contact circle along ABC until the sphere spontaneously detaches from the substrate or “pull-off” at C with a non-zero radius ($a^\dagger > 0$). The “pull-off” at $F^\dagger$ occurs under fixed load when $dF/da \to \infty$ or $da/dF = 0$. The branch CD with $dF/da < 0$ is physically inaccessible under fixed load because $F^\dagger$ is already the minimum force along this curve, but possible under displacement control or fixed grips. The contact shrinks further along CD. At D, the contact is reduced to a point with $a = 0$ and the solid resumes its spherical shape. Along DO, the sphere is readily out of intimate contact with the substrate, but the meniscus turns into a water bridge still exerting a traction on the sphere. Increasing tension reduces the bridge width until it breaks at O, the sphere then completely breaks free from the substrate at $F^{\dagger\dagger} = 0$. Point O is coined “pull-off” under fixed grips. The detachment trajectory is similar for other moisture level. In the dry conditions with $RH \approx 0\%$, $r_k \to 0$, $p^* \to \infty$, the cohesive zone vanishes and a large pull-off force $|F^\dagger|$ is predicted due to a constant $\gamma$. As $RH$ increases, $|F^\dagger|$ approaches the limit of $F^\dagger = -2\pi R \gamma$ along CD (gray curve) and $a^\dagger$ approaches zero, which are summarized in the Figure 3.3. It is noted that the model fails when $2r_k$ is of the same order of magnitude as $R$. 
Figure 3.3. Adhesion of an elastic sphere, with elastic modulus of 1GPa, in moist air. (a) Contact radius as a function of applied load for RH = 5%, 15%, 25%, 35%, 45%, 65%, 85%, and 95% based on the present model (dark curves). At RH = 5%, external tension shrinks the contact area along ABCDO and causes “pull-off” under fixed load at C. Fixed grips continues along CD. At D, a point contact is left and “pull-off” occurs at O. The gray curve shows the locus of fixed load “pull-off”. The Bradley’s
model is essentially the horizontal axis since contact radius is always zero. The DMT model (red dashed curve) is based on the surface tension of water and “pull-off” occurs at D. The JKR model (blue dashed curve) shows “pull-off” under fixed load at P and “pull-off” under fixed grips at P’. (b) Applied load as a function of approach distance for \(RH = 5\%, \ 25\%, \ 45\%, \ 65\%, \ 85\%, \ \text{and} \ 95\%\) based on the present model (dark curves). Detachment proceeds along CDH. “Pull-off” under fixed load occurs at C and under fixed grips at H. The “pull-off” locus are shown as gray curves, along with the DMT and JKR models. The Bradley’s model is identical to the present model under external tension.

Figure 3.3 (b) shows \(F(\delta)\) for a range of \(RH\). For \(RH = 5\%\), \(\delta\) is non-zero at \(F = 0\) due to adhesion (not shown). Applied tension \((F < 0)\) further reduces \(\delta\) until “pull-off” under fixed load occurs at C with maximum tensile load with \(dF/d\delta = 0\). Under fixed grips, \(\delta\) can decrease further under stable equilibrium. At D, \(\delta = 0\) and the sphere resumes its spherical shape. Here the cohesive zone extends to the meniscus with \(a_1 = 2 \ (R\cdot r_k)^{1/2}\), and the external force to maintain equilibrium becomes \(F = -F_{ad} = -p^* \times \pi \cdot a_1^2 = -2\pi R\gamma\). Further decrease in \(\delta\) \((< 0)\) detaches the sphere from the substrate, but the sphere is linked by a bridge to the substrate. To maintain mechanical equilibrium along DH, the applied load is balanced by the adhesion force,

\[
F = -F_{ad} = -p^* \times \pi \cdot a_1^2 = -2\pi R\gamma \times \left(1 + \frac{\delta}{2r_k}\right)
\]

where \(a_1 = \left[2R\cdot (\delta + 2r_k)\right]^{1/2}\) is the bridge radius. As the sphere moves further out from the substrate, the water bridge shrinks and the external load decreases linearly with \(\delta\).

At H, \(\delta^{\dagger\dagger} = -2r_k\), \(a_1 = 0\), the meniscus bridge collapses, the applied tension vanishes, and “pull-off” occurs at \(F^{\dagger\dagger} = 0\). The fixed grips “pull-off” locus is along the negative \(\delta\)-axis, corresponding to O in Figure 3.3 (a). At high \(RH\), \(F^{\dagger\dagger} \rightarrow -2\pi R\gamma\) and \(\delta^{\dagger\dagger} \rightarrow -\infty\).

Figure 3.4 shows \(F^{\dagger}\), \(a^{\dagger}\), and \(\delta^{\dagger}\) as functions of \(RH\). In Figure 3.4 (a), the calculated \(F^{\dagger}(a=0, \delta=0)\) slightly deviates from the DMT limit of \(-2\pi R\gamma\) at very high \(RH\) as the cohesive zone now extends to the sphere dimension \((a_1 \approx R)\) and the Hertz assumption breaks down. Figure 3.4 (b) shows the expected monotonic decreasing function of \(a^{\dagger}(RH)\) that approaches \(a^{\dagger} = 0\) in saturated moisture. Figure 3.4 (c) shows \(\delta^{\dagger}\) \((> 0)\) under fixed load and \(\delta^{\dagger\dagger}\) \((< 0)\) under fixed grips as \(RH\) varies. A drastic decrease in \(\delta^{\dagger\dagger}\) is expected as \(RH\) exceeds 50% since the large meniscus \((r_k)\) sustains a long water bridge (c.f. Eq (3-9)).
Figure 3.4. “Pull-off” as a function of relative humidity: (a) applied tension, $-F^\dagger(RH)$, and (b) contact radius $a^\dagger(RH)$, under fixed load (c.f. gray curve CD in the Figure 3.3 (a)); and (c) approach distance, $\delta^\dagger(RH)$, under fixed load (c.f. gray curve CD in the Figure 3.3 (b)) and fixed grips (c.f. gray curve of negative horizontal axis in the Figure 3.3 (b)).
3.4 Discussion

The new model is rigorously compared with the classical adhesion models: (i) Bradley’s model (Kendall, 2001) where the sphere is rigid and non-deformable, (ii) Derjaguin-Muller-Toporov (DMT) theory (B. V. Derjaguin et al., 1975) for small but stiff spheres in the presence of a weak force with an ideal infinite range, and (iii) the Johnson-Kendall-Roberts (JKR) theory (K. L. Johnson, K. Kendall, & A. D. Roberts, 1971) for large but soft spheres in the presence of a strong surface attraction with an ideal zero range.

In the Bradley’s model, a rigid sphere makes only a single point contact with the substrate with $\delta = 0$ and $a = 0$ independent of both $RH$ and $F$. In the presence of a meniscus, the sum of the applied load and Laplace pressure is balanced by the reaction force at the point contact in the form of a delta function such that the adhesion force is a constant with $F_{ad} = 2\pi R\gamma$ for $F \geq 0$. When $\delta$ turns negative, the sphere is out of intimate contact with the substrate but is linked by a water pillar. Equation (3-11) holds and the behavior is identical to the present model. Pull-off occurs when the water pillar collapses.

In the classical JKR model (Maugis, 2000), the disjoining pressure is taken to be infinite with a vanishingly small force range though the energy to bring the two adhering surfaces together is finite. The tensile stress at the contact edge is therefore theoretically infinite and is capable of deforming the sufficiently soft sphere to the Griffith parabola with $\partial y/\partial r\big|_{r=a} \rightarrow \infty$. An energy balance predicts “pull-off” under fixed load at $P$ in Figure 3.3, where $F^+ = -(3/2)\pi R\gamma$, $a^+ = [9\pi R^2\gamma (1-\nu^2) / 4E]^{1/3}$, and $\delta^+ = - (a^+)^2 / 3R$, whereas “pull-off” under fixed grips occurs at $P'$ where $F'^+ = -(5/6)\pi R\gamma$, $a'^+ = [\pi R^2\gamma (1-\nu^2) / 8E]^{1/3}$, and $\delta'^+ = -3(a'^+)^2 / R$. A close examination of these underlying assumptions prompts one to check for limitations. Laplace pressure cannot increase indefinitely but has an upper bound of the hydrogen bonds of water in the molecular scale or vapor pressure in the macroscopic scale. It is unable to deform a sufficiently stiff sphere (e.g. silica glass) to the ideal parabolic shape at the contact edge, since hydrogen bonds will be broken and water molecules dissociate beforehand. The present model predicts $a^+ > 0$ under fixed load, but there is no local deformation at the contact edge as in JKR. Both $F^+$ and $a^+$ are distinctly different from the JKR values.

The present model is similar to the DMT model to some extent, in that, the sphere deforms according to the Hertz theory and the adhesion mechanics serves as the...
limit for RH approaching saturation as shown in Figure 3.3 (b). Maugis’s version of DMT assumes an infinite force range (D. Maugis, 1992) and \( a_1 \to \infty \). “Pull-off” under fixed grips occurs at \( \delta \to \infty \), and the external tension to maintain the infinite water pillar remains constant at \( F^\dagger = -2\pi R\gamma \) for all \( \delta \leq 0 \). In the absence of sphere-substrate interaction, the “crack front” is located literally at the meniscus, which is consistent with the classical experiment in measuring surface tension of a liquid trapped between two cantilevers (Wan & Lawn, 1990; Wan et al., 1992). One interesting implication of DMT is that the water molecules must penetrate all the way from the meniscus \( z(r=a_1) = 2r_1 \) to the contact edge at \( z(r=a) = z_0 \) so that the Laplace pressure is present in the entire stretch of the cohesive zone. In reality, the physical dimension of two water molecules bonded by a typical hydrogen bond is roughly 8.4Å, as shown in the Figure 3.5 (a). A vacuum thus exists between the contact edge and the molecular wedge, or, \( p = 0 \) in \( r(z_0) \) to \( r(z = 8.4\text{Å}) \). In the limit of \( RH = 100\% \), \( a_1 \gg r(z = 8.4\text{Å}) \), and our model approaches DMT.
Figure 3.5. (a) Schematic of contact edge at molecular level. Symbols of circle filled with white represent molecules of objects in contact. Dark and grey circle represent water molecules. (b) Deformed profile at the contact edge exposed to moist air in the absence of an applied load $F = 0$. Minimum thickness of liquid water is set to be 8.4 Å (dashed line). At RH = 40%, the profile changes abruptly at the circle marked, indicating the meniscus or cohesive zone edge. The JKR and DMT limits are shown as dashed curves.
In the intermediate range of RH, the new model requires the Laplace pressure to be bounded by the cohesive zone, $p = p^*$ in $r(z = 8.4\text{Å})$ to $r(z = 2r_k)$. Based on Eq(3-9), $2r_k = 8.4\text{Å}$ at $RH \approx 30\%$, and no meniscus can exist below this critical RH. The corresponding Laplace pressure is too weak to deform the contact edge to the Griffith parabola. The contact edge can now be arbitrarily chosen to be in the small range between $r(z_0)$ to $r(z = 8.4\text{Å})$. Without loss of generality, we choose to assume the Hertzian compressive stress within the contact circle and the Laplace pressure without, regardless the exact location of the contact edge. To further justify the new model, Figure 3.5 (b) shows the deformed profile for $E = 10$ GPa and a range of RH according to Maugis’s cohesive zone model along with the JKR and DMT limits. All curves show a characteristic kink where the slope $\partial z/\partial r$ changes abruptly indicating the cohesive zone edge $r = a_1$. At $RH < 30\%$, $2r_k < 8.4\text{Å}$ and inter-surface force does not exist. Therefore, meniscus alone is unable to lead to the JKR limit. In Maugis’s calculation, the compressive contact stress gradually turns tensile radially outwards and reaches the maximum tension matching the Laplace pressure. However, the absence of intrinsic solid-solid interaction and the finite water molecular dimension do not allow tension within the contact. Our model allows a discontinuity in stress at the contact edge where it abruptly drops from zero based on Hertz to the full strength of Laplace pressure.
Figure 3.6. “Pull-off” force under fixed load as a function of relative humidity, showing the present model, theoretical results from References (Fogden & White, 1990) and (Xue & Polycarpou, 2008), and experimental data from References (Christenson, 1988). Data for RH < 30% are invalid in the present model and are linked by dashed curve.
It is worthwhile to mention that Christenson (Christenson, 1988) measured an increasing pull-off force as $RH$ rises as shown in Figure 3.6. The ostensible contradiction seems to be explainable by the van der Waals attraction at the solid-solid interface that overshadows the Laplace pressure. Polycarpou (Xue & Polycarpou, 2008) used a numerical approach to show $F^\dagger$ as a monotonic decreasing function of $RH > 70\%$ that approaches the DMT limit at saturated moisture. Such results match with the present model quite well, though Polycarpou did not consider size of water molecules. One major outcome of the current work is to decouple the capillary force from the solid-solid interaction. In the presence of a strong sphere-substrate interaction, the capillary force becomes negligible and the adhesion-detachment mechanics approaches the JKR limit, which is the beyond the scope of this study.

As a final remark, implications of the new model are significant. For instance, it is known that heating and drying helps preventing coagulation in powder (e.g. glass beads, dry spray) and stiction in micro-devices. The present work shows that once $RH$ drops to roughly 30%, water condensation and the consequent adhesion in glass become relatively unimportant.

3.5 Conclusion

A simple adhesion model is derived for an elastic sphere adhering to a rigid planar substrate in the presence of moisture. The model is useful in discussing adhesion of microscopic objects, and the derived values of “pull-off” force and contact radius can be used to deduce the magnitude and range of intersurface forces. Contrary to the classical description based on Maugis’s JKR-DMT transition, meniscus alone causes $|F^\dagger|$ to exceed $2\pi R \gamma$ for any relative humidity and the DMT limit only holds at saturation.

3.6 Acknowledgements

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Chapter 4. Coupled effect of flow rate, ionic strength and bacterial type on bacterial filtration in column test

In this Chapter, the effects of flow velocity and impact of static mechanical properties of bacterium on their attachment and transportation are studied. It is expected that such correlation would vary as a function of liquid flow, aspect ratio of bacterium to collector and a number of relevant parameters related to bacterial structure and aqueous environment. In this chapter, we focus on the effect of flow rate and ionic strength on bacterial filtration. We conducted handful column tests on three different types of bacterium, Aeromonas punctate (strain Q), Bacillus cereus (strain H) and Raoultella ornithinolytica (strain A), under varying flow rates from 5 to 50 mL/min (filtration velocity 0.015 to 0.15 cm/s), varying ionic strengths of 1, 3 and 10 mM. The range of experimental parameters in this study covered the previous test parameters and thus we compared the two for validation purpose. The correlation between μ and α, as defined in Section 1.1.5, was further studied under higher flow rate. A theoretical analysis based on moment balance between fluid drag and adhesion was carried out and showed a great potential of interpreting how flow velocity affects bacterial detachment under varying Derjaguin, Landau and Verwey, Overbeek (DLVO) interaction.

4.1 Materials and Method

4.1.1 Bacterial preparation

Three bacterial strains to be investigated have relevance related to environment and human health (Y. Li et al., 2014). Special features are listed as follows:

(i) Q: *Aeromonas punctata* (Gram-negative) is reported to be associated with human diseases including gastroenteritis, cellulitis and diarrheal

(ii) A: *Raoultella ornithinolytica* (Gram-negative) is found to be a major cause of histamine fish poisoning

(iii) H: *Bacillus cereus* (Gram-negative) is common culprit in food poisoning, causing both intoxication and infections.

All of these strains are aerobic and were grown aerobically at 37 °C in 25 g/L Luria-Bertani (LB) medium (Sigam-Aldrich, Inc., St. Louis, MO). The materials and surface properties of bacterium can be found in reference (Y. Li et al., 2014). To eliminate the potential impact of the growth stage on bacterial micro adhesion properties, all sample cells are in stationary growth phase, namely 16-18 hours growth.
4.1.2 Experiment procedure

Figure 4.1 shows the schematic of flow-through saturated sand packed column test. Ultrapure silica sand GRANUSIL 4095 (UNIMIN Corp., LeSueur, MN) with a nominal diameter of 0.289 mm was utilized as collector in the column testing. The sand was cleaned with 1 M NaOH for 24 hours, rinsed with DI water, dried in an oven at 103 °C for 24 hours, followed by drying in a 550 °C oven for 1 hour before use. A suspension of bacteria was pumped through a 60 mL sterile syringe (i.d. of 2.67 cm) packed with clean silica sand to a height of 10 cm (Y. Li et al., 2014). A column packing porosity of 0.4 was determined by applying standard gravimetric methods (Y. Li et al., 2014). The total pore volume of a packed column is obtained by 60 mL×0.4 = 24 mL. This is defined as pore volume, or PV. 20 pore volumes of DI water followed by 10 pore volumes of the background electrolyte solution (1, 3, 10 mM Potassium Chloride solution) were pumped through the syringe at a certain flow rate, ranging from 5, 10,
20, 30, 40 and 50 mL/min (filtration velocity of 0.015, 0.03, 0.06, 0.09, 0.12, 0.15 cm/s), before each test to make sure the packed column was equilibrated. A suspension of bacteria in the same background electrolyte solution was pumped for 3-4 pore volumes (PV, 1 PV=24 ml), followed by bacteria-free background electrolyte solution (about 3 pore volumes) at the same rate as velocity used in previous step. To make sure the bacteria concentration is constant during the test, a miniature magnetic stir bar was applied in the bacteria solution tank and the bacterial concentration at the inlet of column was measured at the beginning and the end of duration of pumping bacteria solution. The bacteria concentration at the column outlet was monitored continuously using optical density measurements at 500 nm with a UV-vis spectrophotometer (model UV Mini 1240 Shimadzu, Kyoto, Japan) and a 1 cm flow through cell. Each test was conducted in triplicate.

4.1.3 Filtration Efficiency (CFT)

Filtration efficiency, or attachment efficiency (Y. Li et al., 2014), is defined as the ratio of experimental single-collector removal efficiency ($\eta$) to the theoretical single-collector contact efficiency ($\eta_0$), i.e., $\alpha = \eta/\eta_0$. Value of $\eta_0$ for each strain is determined using the expression proposed by Tufenkji and Elimelech as:

$$\eta_0 = 2.4A_s^{1/3}N_R^{-0.081}N_{Pe}^{-0.715}N_{vdW}^{0.052} + 0.55A_s N_R^{1.675}N_{Pe}^{-0.125} + 0.22N_R^{-0.24}N_G^{1.11}N_{vdW}^{0.053}$$

(4-1)

The first term is due to the transport by diffusion, the second term is due to the transport by interception and the last term is the transport due to gravity. Note that Elimelech included the hydrodynamic interaction in this expression. In our study, the gravity term is neglected since the density of bacteria is similar to water.

The single-collector removal efficiency can be determined from the column test breakthrough curve as follows:

$$\eta = \frac{-2d_c \ln(1 - C/C_0)}{3(1 - \varepsilon)L_0}$$

(4-2)

where $d_c$ is the diameter of the quartz sand, $\varepsilon$ is the bed porosity, and $L_0$ is the packed column length. The normalized column effluent concentration, $C/C_0$, was obtained from each breakthrough curve by averaging the values measured between 1.8 and 2 (i.e., clean bed condition). Filtration efficiency $\alpha$ based on the breakthrough curve is then determined as $\alpha = \eta/\eta_0$. 
4.1.4 Bacterial properties derived from AFM (Modified Tabor’s Parameter)

The modified Tabor’s parameter was introduced in section 1.1.4.3, as follows:

\[
\mu = \left[ \frac{2b_2}{l^3E^2} \left( \frac{U_{ad}}{\pi R_{AFM}} \right)^2 \left(1 - \nu^2\right)^{\frac{1}{2}} \right]^{\frac{1}{3}}
\] (4-3)

where \(b_2\) is the minor axis of the cell, \(l\) is the thickness of the cellular surface substances (CSS), \(E\) is the cell elastic modulus, \(U_{ad}\) is the total adhesion energy to detach the AFM tip from sample cell, \(R_{AFM}\) is the AFM tip radius, and \(\nu\) is the Poisson’s ratio. Detailed descriptions of the assumptions made and computation methods for deriving Tabor’s value can be found in reference (Y. Li et al., 2014). It is anticipated that large \(\mu\) facilitates the compliant cells to adhere. It is therefore logically expected that a larger \(\mu\) leads to higher deposition rate in porous medium. The correlation between \(\mu\) and bacterial filtration efficiency \(\alpha\) was introduced in Section 1.1.4.3. In this chapter, we will investigate the correlation between \(\mu\) and \(\alpha\) under varying flow rates and ionic strengths.

4.2 Results and Discussion

4.2.1 Breakthrough curves

Normalized bacterial concentration at the outlet of the column as a function of injected volume of test solution, known as breakthrough curve, were plotted for three strains. Figure 4.2 shows typical breakthrough curves for strain A suspension in 10 mM KCl with flow rates of 5 to 50 mL/min. In the breakthrough curve, the bacterial efflux concentration \((C/C_0)\) is plotted as a function of pore volume passing through the column. Initially, the column is stabilized by 20 pore volumes DI water followed by 10 pore volumes bacterial-free KCl with desirable strength (1 mM, 3 mM or 10 mM). A bacterial suspension in KCl electrolyte is pumped into packed sand column at PV = 0 with flow rates of 5 to 50 mL/min (arrow A). After approximately 1 PV, the injected bacterium penetrated the column and were detected at the outlet (arrow B). As pumped volume increasing, more bacterial cells were monitored at column outlet and the bacterial efflux concentration dramatically increases along BC. The filtration efficiency of each packed column evaluated by the averaging \(C/C_0\) values of breakthrough curve between PV of 1.8 ~ 2.0, as shown in the shadow area of Figure 4.2 pointed by arrow C. The lower the flow rate of bacterial suspension is, the more likely the bacteria are
removed from medium and trapped onto the sand surface, resulting in lower bacterial efflux concentration detected at column outlet. For example, the value of $C/C_0$ is approximately $0.95$ at arrow C for flow rate of 50 mL/min, indicating only 5% of the total bacterium pumping into the column are retained on the sand surface due to adhesion. The value of $C/C_0$ is approximately $0.45$ at arrow C for flow rate of 5 mL/min, indicating as much as 55% of bacterium pumping into the column are trapped in the medium. When attachment equilibrium is established between bacterial cells and sand, breakthrough curve reached plateau. It took longer for the system reaching equilibrium state under high flow rate, e.g. attachment equilibrium was reached at around 2 PV under flow rates of 30 – 40 mL/min. When value of $C/C_0$ reached 100%, the medium in the column was saturated and it could not trap more bacterium from this point on. The column was saturated at ~3 PV for high flow rate of 30 to 50 mL/min. For low flow rates of 5 to 20 mL/min, the column was not saturated for the whole test. At 4 PV (arrow D), the influx is switched to a bacterial free KCl solution with same concentration as bacterial suspension. The bacterial concentration in the effluent increase a little bit along DE for about 1 PV and then decreases abruptly. The slight increase is believed due to residual bacterial suspension in the column and detachment of weak bacterial cell-sand attachment after influx switching. After around 10 PV, there was no bacterial cells detected from the efflux, indicating that the bacterial suspension in the packed column was totally replaced by KCl solution.

Figure 4.3 shows the influence of ionic strength on strain A under two extreme flow rates tested of 5 and 50 mL/min. According to DLVO theory, the increase of ionic strength in KCl electrolyte results in increase of surface charge density and thus the surface charge potential, causing a reduction in the repulsive electrostatic double layer forces. This drives higher ratio of bacterium achieving a strong attachment on sand surface, so called primary minimum(Y. Li et al., 2014; Tufenkji & Elimelech, 2004b; X. Wang et al., 2012). Figure 4.3 (a) demonstrates that more bacterium achieved strong attachment on sand surface at high ionic strength (10 mM KCl), and less bacterium traveled through the column and was detected. However, under high flow rate of 50 mL/min, such a distinct behavior was not observed and the medium reached saturation very fast, as shown in Figure 4.3 (b). The coupled effect of ionic strength and flow rate on filtration behavior was implied by the test observations.
Figure 4.2 Breakthrough curve of strain A under ionic strength of 10mM, with a wide range of flow rates (5, 10, 20, 30, 40 and 50 mL/min)
Bacterial efflux concentration, $C/C_0$

PV (1 PV=24 ml)
Figure 4.3 (a) Breakthrough curve of strain A under flow rate of 5 mL/min, with a wide range of ionic strengths (1, 3 and 10 mM KCl). (b) Breakthrough curve of strain A under flow rate of 50 mL/min, with a wide range of ionic strengths (1, 3 and 10 mM KCl)
Figure 4.4 Breakthrough curves for strain A, Q and H with ionic strengths of 1, 3, and 10mM and flow rates of 5, 10, 20, 30, 40 and 50 mL/min. Different colors of red, yellow and blue are applied here to indicate different ionic strength. A single chart indicated breakthrough curves for one type of bacterium at constant ionic strength and varying flow rate. For example, subplot (a) indicated breakthrough curves of strain A under ionic strength of 1mM and flow rate of 5, 10, 20, 30, 40 and 50 mL/min. The curve at the bottom corresponded to test under lower of flow rate, and vice versa.
Overall breakthrough curves for all three strains under all experimental parameters were shown in Figure 4.4. Filtration efficiency of different bacterial strains were previously studied and was ranked as A<Q<H (Y. Li et al., 2014). A similar behavior was observed in this study, as shown in Figure 4.4. Other than strain Q is “stickier” than strain A, the pattern of breakthrough curve for strain Q is similar with strain A. For example, strain Q never reached saturation under 10 mM KCl. Strain H is most “sticky” and the column was not saturated under any conditions. Based on the breakthrough curves, the filtration efficiency α was derived, as described in Section 4.1.3, which provides a clear way to investigate the effects of flow rate, ionic strength and bacterial species on filtration efficiency of the system.

4.2.2 Filtration Efficiency α

Filtration efficiency α for three bacterial strains under varying flow rates and ionic strengths are shown in Figure 4.5. We observe that α decreases as flow rate increases for both strain A and Q under all ionic strengths as indicated in Figure 4.5(a) and (b). This can be explained by impact of hydrodynamic interaction on two filtration mechanisms, namely surface adsorption and clogging. On one hand, the hydrodynamic drag force exerted on adhered bacteria increases as flow rate increases. This results in a lower attachment ratio of bacterium on a collector surface. On the other hand, the clogging was broken through under higher hydrodynamic pressure. Since the ratio of bacterium size to sand size is smaller than 0.05, clogging is not dominant in our column test, as explained in Section 1.1.4.2. The detachment of bacterium from the collector surface due to hydrodynamic interaction is the cause of this inverse relationship between the filtration efficiency and the flow rate. Note that strain H has a high modified Tabor’s parameter μ. These bacteria were able to achieve strong attachment and flow rate had no effect on their detachment, as indicated by Figure 4.5 (c).

Filtration efficiency α also shows a positive correlation with ionic concentration for strain A and strain Q, indicated by Figure 4.5(a) and (b). As ionic concentration increases, more bacteria are removed by the porous medium system. This can be explained by the classic DLVO theory, as demonstrated in Section 2.3. Bacteria in the 1st min experiences strong adhesion while bacteria in the 2nd min experience weak adhesion. When ionic concentration is low, the deposition is in an unfavorable mode with a relatively high repulsive barrier. Very few bacteria overcome the barrier and go into the 1st min. At the same time, low ionic strength also reduces the total adhesion
energy in the 2\textsuperscript{nd} min. When ionic concentration is high, the EDL repulsion is decreased and van der Waals (vdW) attraction dominates. Most or all bacteria go into the 1\textsuperscript{st} min, defined as the favorable deposition mode. Adhesion energy in the 2\textsuperscript{nd} min is also increased. In summary, high ionic strength results in higher ratio of bacterium adhering in the 1\textsuperscript{st} min and stronger attraction in 2\textsuperscript{nd} min. Low ionic strength results in smaller ratio of bacterium adhering in the 1\textsuperscript{st} min and weaker attraction in 2\textsuperscript{nd} min. Strain H is an exception due to its strong surface adhesion energy.
Figure 4.5 Filtration efficiency, or attachment efficiency $\alpha$, derived from column test, as a function of flow rate.
4.2.3 Modified Tabor’s parameter

Li et al. concluded a strong correlation between micro properties of bacterium and their macro filtration behaviour at given flow rate of 5 mL/min and ionic strength of 1, 3 and 10 mM, as discussed in section 1.1.4.3 (Y. Li et al., 2014). In this study, under the same experimental conditions, the correlation was observed again and it agreed very well with the previous results derived by Li, as indicated by red and blue symbols in Figure 4.6. The pink symbols in Figure 4.6 represent the filtration efficiency $\alpha$ of three strains under 50 mL/min versus their values $\mu$. The new correlation shifted, as implied by the pink dash line in Figure 4.6. Note that effect of flow rate on strain H is negligible.

![Figure 4.6](image)

Figure 4.6 Correlation between modified Tabor’s parameter, $\mu$ based on AFM measurement and filtration efficiency, $\alpha$ based on column test, under two different flow rates of 5 and 50 mL/min.

Bacteria with low value of $\mu$ are rigid and of small adhesion energy and they are more sensitive to drag force due to flow. As shown in the Figure 4.6, $\alpha$ of strain A scattered in the vertical direction as flow rate increases from 5 to 50 mL/min. Bacteria with high
value of $\mu$ are typically soft and of strong adhesion energy. When attached on the collector surface, they are compliant and difficult to be detached in the presence of flow.

4.3 Conclusion

In this chapter, we conducted handful column tests on three bacterial strains that are related to environmental pollution and human health. We investigated the impact of flow rate and ionic strength on bacterial filtration efficiency. For strain A and strain Q, we found a inverse relationship between the filtration efficiency and the flow rate. At the same time, we found a positive correlation between filtration efficiency and ionic strength. Based on the DLVO theory, we conducted a qualitative analysis on surface interaction between bacterium and collector surface under varying ionic strength. Coupled effect of ionic strength and flow rate were implied by the breakthrough curves of strain A and strain Q.

The modified Tabor’s parameter, $\mu$, captured the significance of single bacterial properties in bacterial filtration process in the porous medium. Yet the modified Tabor’s parameter did not consider the flow condition in this process. Both original Tabor’s parameter and modified Tabor’s parameter are demonstrating a static characterization of subject, e.g. bacterium in this study. In this chapter, we observed apparent influences of flow rate and ionic strengths on bacterial filtration. This further proved that bacterial filtration in the porous medium is a complex mechanical/biological process which requires knowledge of flow condition, bacterial/collector properties and aqueous environment. In the following chapters, we would use a new experimental method on bacterial filtration and investigate the coupled the effect of ionic strength and flow rate on bacteria detachment.
Chapter 5. Measuring Particle Adhesion in the Presence of DLVO Potential by Microfluidics

As the mechanical properties of the particles/pathogens (Y. Li et al., 2014; X. Wang et al., 2012) and the environmental conditions (Scott A. Bradford, Torkzaban, Leij, Šimůnek, & van Genuchten, 2009; Brown et al., 2002; J. Li, Xie, & Ghoshal, 2015; Shen et al., 2010; Torkzaban et al., 2007) were shown to affect their filtration behaviour, the conventional column test showed limitations in delineating the effects of individual factors and incorporating new theoretical models (J. Li et al., 2015; X. Li, Zhang, Lin, & Johnson, 2005; Shen et al., 2010; Torkzaban et al., 2007). In this chapter, a new experimental approach is introduced. The new test method excluded the particle/pathogen transport from the problem and merely concentrated on the particle/pathogen adhesion-detachment mechanics in the presence of flow. The inter-surface force and hydrodynamic interaction were calculated. An empirical equation for filtration efficiency $\alpha$ was suggested. Lastly, a theoretical model based on moment balance is adopted and theoretical results were compared with the experimental results.

5.1 Introduction

Filtration is widely used to remove microbes, colloids, and particles from polluted water. The conventional way of gauging the filtration efficacy of a medium is to flow a particle rich solution through a sand column while monitoring the number of particles retained as a function of time, which in turn depends on the ionic concentration of the solution, flowrate, temperature, etc., as shown in Figure 1.3. To better assess the fate of particle transport, a widely accepted theoretical model based on the underlying convective-diffusion equation (1-2) is available in the literature (Friedlander, 1958; van Smoluchowski, 1917). Yao et al. (Yao et al., 1971) presented the classical semi-empirical colloid filtration theory (CFT) that was further improved by Rajagopalan (Rajagopalan & Tien, 1976) and Elimelech (Elimelech, 1994; Tufenkji & Elimelech, 2004a). Other methods include subjecting the adhered particles to increasing hydrodynamic shear, and monitoring when the particles detach from the substrate (Sharma, Gibcus, van der Mei, & Busscher, 2005; Shen et al., 2010; Torkzaban et al., 2007). Xin et al. demonstrated how the macro-scale filtration efficiency was related to the microscopic properties of individual bacterial cells (Y. Li et al., 2014; X. Wang et al., 2012). The cell-substrate adhesion and elastic modulus of the cell wall were measured using an atomic force microscope (AFM), and the cell geometry and
dimension by optical microscope. A dimensionless Tabor’s parameter comprising these intrinsic cell properties was shown to be correlated with the macroscopic filtration efficiency (Y. Li et al., 2014; X. Wang et al., 2012).

Despite the fact that the classical CFT theory incorporates the impact of flow rate and ionic concentration on filtration via the convective-diffusion equation and filtration test, the underlying mechanisms behind these two factors are not clear. For example, several trapping mechanisms coexist in the traditional filtration test, namely, physical straining, aggregation, and surface attachment (W. P. Johnson, Tong, & Li, 2007; Shen et al., 2010; Tong & Johnson, 2006; Torkzaban et al., 2015; Zhang, Raoof, & Hassanizadeh, 2015). These could affect particle detachment via: (i) high flow rate that disrupts any clogging caused by colloidal/bacterial aggregation (Bergendahl & Grasso, 2000; X. Li et al., 2005; Shen et al., 2010; Torkzaban et al., 2015; Torkzaban et al., 2007) and (ii) high hydrodynamic drag force that detaches the colloids/bacteria adhered to the collector surface (Tong & Johnson, 2006). In this chapter, we introduce a new microfluidic device based method to exclude the effect of straining and aggregation, but focus on particle-collector intersurface interaction and the coupled influence of flow rate and ionic concentration (W. P. Johnson et al., 2007; Shen et al., 2010; Tong & Johnson, 2006). One advantage of such approach is the logical extension of modeled polystyrene particles to bacterial cells in the long run, which is not discussed in the classical CFT. A new mechanical model based on experimental findings will therefore provide a practical way to assess colloidal attachment efficacy. A new microfluidic device is designed, constructed and tested to investigate particle attachment/detachment. Both surface chemistry and flow dynamics are incorporated into colloidal filtration model based on the classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory (Elimelech & O’Melia, 1990; Hogg et al., 1966) and rudimentary fluid mechanics.

5.2 Material and Methods

5.2.1 Microfluidic Device

A microfluidic channel was fabricated on a quartz petri dish. A Scotch tape™ (3M) was cut to a desired width and adhered to the substrate to create a master for replica molding. Polydimethylsiloxane (PDMS) precursor was mixed at a 1:10 ratio with the curing agent (Sylgard 184; Dow Corning) and poured into the mold before vacuum was pulled to eliminate air pockets. The polymer was then cured at 69˚C for 2
hours. The microchannel had a cross-section of 70 μm × 3.5 mm. Three holes were drilled through the PDMS slab along the micro-channel. The hole at one end served as the inlet and the hole at the other hand the outlet. The inlet at the midspan allowed particle rich solution to be injected into the channel. The set up was then sonicated, further cured in deionized water for 1 hour, cleansed by isopropyl alcohol, subjected to another hour of further sonication, and thoroughly drying by nitrogen. Bonding of PDMS onto a glass microscope slide was enhanced by gently pressing to remove any air pockets, followed by thermal treatment at 85°C for 2 hours. The PDMS-glass interface was sufficiently strong to meet the pressure requirement without leakage. The device was reusable after proper cleaning. Figure 5.1 (a) shows a typical microfluidic channel with the inlet, intermediate inlet, and outlet.
Figure 5.1. (a) The microfluidic device. The arrow indicates the flow direction of background solution. The dashed line indicates the extent particles can reach from the point of injection. (b) Experimental setup.

5.2.2 Experimental Setup and Method

Figure 5.1 (b) shows the experimental setup. The microfluidic device was placed on an inverted optical microscope platform for in-situ observation. Potassium chloride solution KCl (aq) of desirable concentration $c = 3$ to $100$ mM was injected into the channel via Inlet-1 using a syringe pump (model NE-300, New Era Pump System Inc., Farmingdale, NY), and exited via the outlet, as shown in Figure 5.1 (b). Once the channel was filled, the flow halted and the liquid was stagnant. Plain polystyrene spheres (PP-30-10, Spherotech, Inc., Lake Forest, IL) with a diameter of 3.43 μm and density of 1.05 g/cm$^3$ were mixed with KCl (aq) to yield a particle number density of $\sim 7 \times 10^5$ μL$^{-1}$. The particles were carefully injected via inlet 2 at 0.03 mL/h. The incoming volume was carefully controlled at approximately 2-3 μL such that particles essentially reached a maximum observable extent and formed a distinct boundary at the upstream location indicated by the dashed curve in Figure 5.1 (a). The particles were
allowed to settle and naturally adhere to the glass substrate or collector, while the hydraulic pressure at the closed Inlet-2 was maintained. An optical micrograph was taken at vicinity of the particle boundary to record the initial particle distribution and areal density on the glass surface. The observation area was chosen since detached particles could only move out of the observation area. If an area close to the injection inlet was chosen, detached particles from the upstream would interfere with the observation. Flow from Inlet-1 at the desired flowrate, $Q$, from 0.03 to 4.0 mL/h then began. Figure 5.2 shows micrographs taken before and after the onset of liquid flow. Micrographs of each test were processed and the number of particles retained on the glass substrate were counted by standard image processing tools in MATLAB (version R2015b, MathWorks, Natick, MA).
Figure 5.2. Particle distribution (O) right after injection and deposition on the glass substrate, and (A, A’, B and B’) after the channel is flushed with background solution of 10 mM KCl (aq) at 0.03, 0.06, 0.3 and 0.6 mL/h.
5.3 Results

The filtration or attachment efficiency, $0 \leq \alpha \leq 1$, is defined as the fraction of retained particles such that $\alpha = 1$ when all particles are trapped, and $\alpha = 0$ when all particles are detached and freely pass through the channel. The time-dependent $\alpha$ is influenced by both $c$ and $Q$, and can be expressed mathematically, $\alpha = \alpha_s(c, V, t)$, with the saturated value of $\alpha_s = \alpha(t \to \infty)$ at specific $c$ and $Q$. Experiments were performed by three methods, and were repeated at least 3 times for specific combination of $c$ and $Q$.

![Figure 5.3](image.png)

Figure 5.3. Number of particles retained on the collector surface as a function of time after volume flowrate increases from 0 to 3.0 mL/h.
Method 1: Constant flowrate. The flowrate was set at a fixed value at $t = 0 \text{ s}$ and held constant. Figure 5.3 shows typical $\alpha(t)$ for $Q = 3.0 \text{ mL/h}$ and a range of $c$. As soon as the flow began, loose particles detached from the glass surface, and $\alpha$ gradually reached $\alpha_s$ within the first 5 seconds. In the solution with $c = 100 \text{ mM}$, virtually all particles remained intact on the substrate and $\alpha \approx 1$. Dilute solution reduced the filtration efficacy until $\alpha_s(3 \text{ mM}, 3.0 \text{ mL/h}) \approx 0$ and virtually all the particles were washed off the microchannel. Intermediate concentrations, $10 \text{ mM} \leq c \leq 30 \text{ mM}$, raised $\alpha$ but significant data scattering was inevitable. Ultralow flowrate of $Q = 0.01 - 10 \mu\text{L/h}$ was investigated in a microchannel with large cross-section of $1 \text{ mm} \times 5 \text{ mm}$ to reduce any minor losses, and images were taken every 2 min. No discernable changes were observed compared to $Q = 0.03 \text{ mL/h}$. Very high flowrates of $Q = 10 - 30 \text{ mL/h}$ were attempted, but no further particle detachment was observed. Thus, $Q_{\text{min}} = 0.03 \text{ mL/h}$ and $Q_{\text{max}} = 3.0 \text{ mL/h}$ were set as the lower and upper limits, respectively, of all measurements hereafter.

Method 2: Stepwise moderate flowrate. Flow was set at $Q = 0.03 \text{ mL/h}$ initially until a steady state was established with a constant $\alpha_s$. Flow was then abruptly increased to $Q = 3.0 \text{ mL/h}$ until a new $\alpha_s$ was reached. Figure 5.4 shows the initial gradual particle detachment and the drastic drop of $\alpha$ due to the sudden increase in $Q$. Values of the new $\alpha_s$ were fairly consistent, with the high $Q$ limits shown in Figure 5.3. Measurements at $10 \text{ mM} \leq c \leq 30 \text{ mM}$ showed significant data scattering.

Method 3: Intermittent increase in flowrate. Flow was set to $Q(t = 0) = 0.03 \text{ mL/h}$ until steady state was reached. The flowrate was then gradually raised to 0.03, 0.06, 0.12, 0.30, 0.60, 1.20, 1.80, 2.40, then 3-4 mL/h. Figure 5.5 shows part of the data from the initial washout. As $Q$ increases, $\alpha$ decreases accordingly. Optical micrographs were collected at one frame per minute. Figure 5.5 shows a typical measurement for 10 mM KCl (aq), from the initial deposition at O, sudden increase in $Q$ from 0.03 to 0.06 mL/h at A-A’, then $Q = 0.6$ to 1.2 mL/h at B-B’. Figure 5.2 shows the images taken at O, A-A’ and B-B’. Less than 50% of particles were left behind at $Q = 0.03 \text{ mL/h}$ at steady state, while abrupt drops of $\alpha$ took place at A-A’ and B-B’.
Figure 5.4. Filtration efficiency as a function of time. Test started at $Q = 0.30 \text{ mL/h}$ and was abruptly raised to $Q = 3.0 \text{ mL/hr}$, leading to a sudden drop in $\alpha$ as indicated by the two arrows for $c = 30 \text{ mM}$. The two dashed curves are curve fits to $c = 10 \text{ mM}$ and $30 \text{ mM}$ to calculate the relaxation time $\tau$. 
Figure 5.5. Filtration efficiency as a function of time. Flowrate was suddenly raised from $Q = 0.3$ to $0.6$ mL/h, then from $Q = 0.6$ to $1.2$ mL/h. In a typical measurement for $c = 10$ mM, the two transitions occurred at AA’ and BB’.
Figure 5.6. Filtration efficiency as a function of concentration, $\alpha(c)$, for flowrate of $Q = 0.30$ mL/h, showing the changes in $\alpha(t)$ for fixed time $t$ after the onset of flow. The gray curve indicates the limiting $\alpha$ at high flowrate $Q^* = 3.0$ mL/h when all particles trapped in 2min are removed.
Data of $\alpha(c,V,t)$ are reorganized in Figure 5.6 and Figure 5.7 to elucidate the filtration behavior. Figure 5.6 shows the characteristic temporal behavior of $\alpha(c, Q = 0.03 \text{ mL/h})$. At any $c$, $\alpha(t = 0) = 1$ at initial deposition (not shown). In concentrated solution with $c = 100 \text{ mM}$, nearly all deposited particles adhered firmly to the substrate and were immobile at any $Q$ and $t$, and therefore $\alpha = 1$. At lower $c$, $\alpha$ became time dependent. A short interval after flow began ($t = 30 \text{ s}$), $\alpha$ diminished until it gradually reached the long time limit of $\alpha_c(c)$ at $t \approx 150 \text{ s}$, beyond which further particle detachment was not observed. The relaxation time from the onset of flow to

![Graph showing fraction of retained particles vs ionic concentration](image)

Figure 5.7. Fraction of particles removed at steady state as a function of ionic concentration ($c = 3 \text{ mM}, 10 \text{ mM}, 20 \text{ mM}, 30 \text{ mM}, 60 \text{ mM}, \text{ and } 100 \text{ mM}$) for a range of flowrate. The gray curve indicates the limiting $\alpha$ at high flowrate $Q^* = 3.0 \text{ mL/h}$ when all particles trapped in 2min are
removed (c.f. Figure 5.6). Data are slightly displaced laterally to avoid interference.

saturation decreases as $c$ increases. As $Q$ increases from 0.03 to 3.0 mL/h (not shown), $\alpha(c)$ shifts downwards and approaches $\alpha_c(Q_{\text{max}})$ at the high flow rate limit (gray curve). Figure 5.7 shows saturated filtration efficiency $\alpha_s(c)$ for a range of flowrates. Once again, the gray curve of $Q = 3.0$ mL/h serves as the lower bound.

### 5.4 Theoretical Model and Data Analysis

**Inter-surface forces**

A model based on classical inter-surface forces and mechanical deformation is developed based on the collected data. In the presence of an electrolyte, interaction between two surfaces is governed by the superimposed van der Waals (vdW) attraction and electrostatic double layer (EDL) repulsion based on the DLVO theory. The interaction energy density between two parallel planar surfaces with a separation of $h$ is given by

$$E_d(h) = -\frac{A_H}{12\pi^2 h^2} + \frac{\varepsilon_0 \varepsilon_r \kappa}{2} \left[ 2\psi_s \psi_p \csc h - (\psi_s^2 + \psi_p^2) \cdot (\coth \kappa h - 1) \right]$$  \hspace{1cm} (5-1)

with Hamaker constant $A_H$, the dielectric constant of the vacuum $\varepsilon_0$, relative dielectric constant of the electrolyte $\varepsilon_r$, zeta potentials of substrate $\psi_s$ and particle $\psi_p$ (Tufenkji & Elimelech, 2004b), and inverse Debye screening length $\kappa$ (Hogg et al., 1966). The potential energy between a sphere with radius of $R$ and a planar substrate surface with a distance of $H_0$ away is

$$U(H_0) = \int_0^\infty E_d(h) \cdot 2\pi r \, dr$$

with $h = H_0 + R - (R^2 - r^2)^{1/2}$. The net force is given by

$$F_{\text{DLVO}}(H_0) = \int_0^\infty \frac{\partial E_d(h)}{\partial h} 2\pi r \cdot dr\bigg|_0^R + \int_a^R \frac{A_H}{6\pi^2 h^3} 2\pi r \cdot dr$$

$$+ \int_a^R \varepsilon_0 \varepsilon_r \kappa (\psi_s^2 + \psi_p^2) \cdot \frac{\partial}{\partial h} \left[ 1 - \coth (\kappa h) + \frac{2\psi_s \psi_p \csc (\kappa h)}{\psi_s^2 + \psi_p^2} \right] 2\pi r \cdot dr$$  \hspace{1cm} (5-2)

Figure 5.8 shows $U(H_0)$, featuring (i) a primary minimum (1min) with strong but short-ranged attraction, (ii) a secondary minimum (2min) with weak but long-ranged attraction, and (iii) a repulsion barrier separating 1min from 2min and ranging from
7500 \( kT \) at 3 mM to -40 \( kT \) at 100 mM. Particles trapped in the shallow 2min energy well are capable to escape from the substrate provided sufficient kinetic or thermal energy is available. In a flow, hydrodynamic shear developed at the liquid-solid boundary leads to detachment and the particle rolls off the substrate. Higher ionic concentration reduces the energy barrier and facilitates the particles to move into the strong 1min where particles are permanently retained. Filtration efficiency depends on the stochastic distribution of particles in 1min and 2min subjected to flow.

**Particle deformation**

When an elastic spherical particle with radius \( R \), elastic modulus \( E \), and Poisson’s ratio \( \nu \) is subject to adhesion, the particle-substrate interface expands from a point to a contact circle with radius \( a \). If the inter-surface interaction is equivalent to an external applied load, deformation of the particle follows the classical Hertz contact theory (Heinrich Hertz, 1896), where the adhesion force, or net force due to the DLVO potential, \( F \), is given by

\[
F_{DLVO} = \frac{4}{3} \left( \frac{E}{1 - \nu^2} \right) \frac{a^3}{R} \tag{5-3}
\]

The deformed geometry is thus related to the environment via the ionic concentration. The probability of a particle being trapped in 1min or 2min can be discussed in terms of a normally distributed, rather than a single-valued, \( \psi_p \). The resulting inter-surface force spans a range and so is the contact radius. In a dilute electrolyte, the energy barrier is so tall that the particles are forbidden to go into the 1st min. Most particles are therefore trapped in 2min and keep a particle-substrate separation equal to the force range rather than being in intimate contact with the substrate. The “pseudo” contact radius is taken to be \( a_2 = [R^2 - (R - z_2)^2]^{1/2} \) by simple geometry, with \( z_2 \) the effective range of 2min.
Figure 5.8. The DLVO potential energy showing (a) repulsive energy barrier and (b) secondary minimum 2min, based on (i) $c = 3 \text{ mM}$, $\psi_p = -80.3 \text{ mV}$ and $\psi_s = 55.2 \text{ mV}$, (ii) $c = 10 \text{ mM}$, $\psi_p = -60.5 \text{ mV}$ and $\psi_s = -50.2 \text{ mV}$, (iii) $c = 30 \text{ mM}$, $\psi_p = -45.3 \text{ mV}$ and $\psi_s = -36.6 \text{ mV}$, and (iv) $c = 100 \text{ mM}$, $\psi_p = -30.1 \text{ mV}$ and $\psi_s = -18.9 \text{ mV}$, and $A_H = 10-20 \text{ J}$. In dilute electrolyte (3 mM), the particles have insufficient energy to surmount the repulsive barrier and stay in 2min. At 100 mM, the energy barrier is reduced to such a level that thermal fluctuation throws the particles into the strongly attractive 1min.
Hydrodynamic drag on adhered particles

Steady flow in the micro-channel follows the Hagen-Poiseuille's equation (Elimelech, 1994) that requires a steady state velocity profile $V(z)$ governed by

$$V(z) = 6 \frac{V_m}{z_0} \left(1 - \frac{z}{z_0} \right)$$

(5-4)

with $V_m$ the mean fluid velocity, $z_0$ the channel depth, and $z$ is the distance from collector surface. Laminar flow is expected as the Reynolds number falls in the range of $Re = 10^{-5} - 10^{-2}$. The hydrodynamic drag is given by (Shen et al., 2010)

$$F_D = 10.205 \pi \tau R^2 \frac{\partial V}{\partial z}$$

(5-5)

with the viscosity of solution $\tau$. Neglecting the lift force on the particle, the hydrodynamic torque is therefore $M_D = F_D \times l$ with $l = 1.4 \times R$ being the moment arm of $F_D$ (Shen et al., 2010). As flowrate increases, $M_D$ reaches the rotational inertia of the adhered particle, the particle pivots at the far end of the contact circle, and eventually breaks loose from the substrate and is carried away by the external flow. A simple criterion can be stated for particle detachment: $M_D > M_D^*$ where the critical torque is given by $M_D^* = F_{Ad} \times a_i$, with $i = 1$ or 2 denoting 1min and 2min. Figure 5.9 shows the hydrodynamic drag as a function of flowrate, along with the detachment thresholds $M_D^*$ of 2min at various $c$. Within the range of $Q$ shown, $M_D(Q)$ spans roughly $10^{-19} - 10^{-17}$ Nm, which overlaps with $M_D^*$ for 2min. Particles are detached once $M_D(Q)$ rises above $M_D^*$. The 1min thresholds are in the order of $\sim 10^{-14}$ Nm that is well above $M_D$. Hydrodynamic drag is therefore unable to detach the particles trapped in 1min. Calculated results of hydrodynamic torque and adhesive torque in 1min and 2min are also shown in the Table 5.1.
Table 5.1. Hydrodynamic Torque (N·m) at flow rate $Q = 0.03 – 3.0$ mL/h and critical torque due to DLVO force in primary and secondary minimum in the presence of 3 to 100 mM KCl (aq). Secondary minimum vanished under ionic concentration of 100 mM thus the critical torque is not applicable.

<table>
<thead>
<tr>
<th>Flow rate (mL/h)</th>
<th>Torque</th>
<th>Torque in 1min (N·m)</th>
<th>Torque in 2min (N·m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.03 ~ 3.0</td>
<td>6.94×10^{-17} ~ 6.94×10^{-19}</td>
<td>I (mM)</td>
<td>1min</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>1.39×10^{-14}</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10</td>
<td>1.00×10^{-14}</td>
</tr>
<tr>
<td></td>
<td></td>
<td>30</td>
<td>1.01×10^{-14}</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100</td>
<td>1.07×10^{-14}</td>
</tr>
</tbody>
</table>

**Empirical equation for filtration efficiency**

Filtration efficiency is understood as an interplay between the 1min and 2min in the presence of external flow. When the particles are first injected into the microchannel and allowed to settle, some are trapped in 1min and the rest in 2min. In a dilute electrolyte, the high energy barrier leaves most particles in 2min which are easily removed even at small $Q$, and $\alpha$ drops to a small value. Conversely, in a concentrated solution, the diminished energy barrier causes most particles to be pulled into the strong 1min and thus remain on the collector surface even at high $Q$, and $\alpha$ approaches unity. The portion in each energy well depends on the height of the energy barrier. Figure 5.5 shows the instant decrease in $\alpha$ due to an intermittent increase in $Q$. As the flowrate exceeds critical value of $Q^* = 3.0$ mL/h, only particles in 1min remain on the substrate. Based on this interpretation, an alternative filtration efficiency, $\alpha_{2\min}$, is defined such that $\alpha_{2\min} = 0$ when all particles in 2min are removed at high $Q$ and $\alpha_{2\min} = 1$ when all particles are retained at low $Q$.

Data shown in Figure 5.6 and Figure 5.7 are now analyzed based on the new model. The electrostatic repulsive barrier in a 3mM electrolyte is $\sim 10^3 k_BT$ with the Boltzmann constant $k_B$ and absolute temperature $T$, which is excessively high compared to the thermal fluctuation of $\sim 1 – 100 k_BT$ at 25°C. Particles are unable to overcome the energy barrier to transit to 1min. Figure 5.7 shows that roughly 3% of particles fall into 1min due to physical and chemical heterogeneities (Elimelech & O'Melia, 1990). As $c$ reaches 60 mM, virtually all particles are trapped in 1min because of the
diminished barrier and cannot be detached by the flow. This also explains why all $\alpha(c)$ curves converge at high $c$. The gray region in Figure 5.7 indicates proportion of particles trapped in 1min, while the clear region corresponds to 2min.

Figure 5.9. Hydrodynamic Torque (N·m) at flow rate $Q = 0.03 - 3.0$ mL/h and critical torque due to DLVO force in primary and secondary minimum in the presence of 3 to 100 mM KCl (aq). Note that secondary minimum vanished under ionic concentration of 100 mM thus the critical torque in 2min is not applicable. Critical torques in 1min under varying ionic concentrations varies in a small range due to van der Waals force dominating adhesion mechanism in this region.
When the particles are first injected into the microchannel, a fraction \( f_{1\text{min}} \) of the particles are trapped by the 1min and the rest, \( f_{2\text{min}} \), by 2min such that \( f_{1\text{min}} + f_{2\text{min}} = 1 \). Apparently, \( f \) depends on the ionic concentration that influences the repulsive barrier in surface potential. Since the particles in 1min are immobile due to strong attraction, \( f_{1\text{min}} \) is time independent and so is its contribution to the overall filtration efficiency. Particles in 2min, however, are detached by liquid flow and its removal rate depends on both \( V \) and \( c \). A simple equation can be written as

\[
\alpha(c, Q, t) = f_{1\text{min}}(c) + f_{2\text{min}}(c) \times \alpha_{2\text{min}}(c, Q, t) \tag{5-6}
\]

where \( f_{1\text{min}} = 0 \) and \( f_{2\text{min}} = 1 \) for \( c \to 0 \), and \( f_{1\text{min}} = 1 \) and \( f_{2\text{min}} = 0 \) for \( c \geq c^* \), with \( c^* \) an experimentally determined threshold. The temporal \( \alpha_{2\text{min}}(c, Q, t) \) governs the gradual particle removal and is taken to be an exponential decay function,

\[
\alpha_{2\text{min}}(C, Q, t) = [ 1 + \alpha_{2\text{s}}(C, Q) \times (e^{t/\lambda} - 1) ] e^{-t/\lambda} \tag{5-7}
\]

where \( \lambda \) is a characteristic relaxation time of particle removal to be determined experimentally. The function \( \alpha_{2\text{min}}(c, Q, t = 0) = 1 \) approaches the saturation limit of \( \alpha_{2\text{min}}(c, Q, t \to \infty) = \alpha_{2\text{s}}(c, Q) \). In Figure 5.6, the high initial removal rate slows down to approach the saturation limit of \( \alpha_{2\text{s}}(c, Q_{\text{max}}) \) shown as the gray curve. Figure 5.7 shows \( f_{1\text{min}}(c) \), or equivalently \( \alpha_{s}(c, Q_{\text{max}}) \), to be the border separating 1min from the complementary 2min. Flow rate only plays a role in the 2min. Figure 5.5 shows that attachment efficiency \( \alpha \) decreases monotonically as a function of flow except under ionic concentration of 100 mM where the secondary minimum disappears. Figure 5.10 shows the monotonic decreasing \( \lambda (c) \) based on curve fitting data in Figure 5.4. Possible reasons of the transient behavior are the finite time to establish steady flow in crevices and surface inhomogeneity where particles are trapped and time needed for interfacial delamination to propagate from the frontal contact edge to the leeside. Further investigation is necessary.
A theoretical model based on moment balance method

The detachment of particles depends on the competition between the hydrodynamic moment and the adhesive moment. If the hydrodynamic moment overcomes the adhesive moment, the particle would be detached and vice versa. Due to physical and chemical heterogeneities and size variation, zeta potential of the particles varies in a range of +/- 15 mV at a given ionic concentration (Bose, Keharia, & Deshpande, 2013; Liu, Zhou, Xu, & Masliyah, 2002). The adhesive moments of particles vary accordingly. Therefore, the fraction of detached particle and flow rate have a positive correlation. In other words, hydrodynamic moment of low flow rate detaches particles with low adhesive moment first. As flow rate increases, the particles with high adhesive moment would be detached. Note that, the adhesive moment calculated here is in the secondary minimum. The variation of particle size is not considered in the calculation of the adhesive moment. We assume that the zeta potential distributed normally with variance of 10 mV. Fraction of particles in the primary minimum is experimentally determined and adopted in the theoretical model. Thus, the filtration efficiency is calculated as a function of flow rate. The theoretical results and experimental results are shown in Figure 5.11.
Figure 5.11 Filtration efficiency calculated by the moment balance method, as indicated by solid lines and measured by the microfluidic test, indicated by the scattered dots.

The theoretical model predicts an abrupt drop of filtration efficiency under different flow rates corresponding to different ionic concentrations. The abrupt drop of filtration efficiency implies that the particles in the secondary minimum are detached rapid due to weak adhesion. On one hand, high ionic concentration suggests higher fraction of particle in the primary minimum. This is indicated by the heights of plateaus of theoretical curves. Note that this is not calculated but empirically derived from the test results. On the other hand, high ionic concentration suggests higher magnitude of adhesion in the secondary minimum. This is indicated by different range of flow rates that causes detachment under different ionic concentrations. The slope of the theoretical curve is determined by multiple factors, e.g. the zeta potential distribution of particles, particle size variation and heterogeneities of glass surface and only the variation of the zeta potential is considered in the present calculation.
5.5 Discussion

Empirical results obtained from the new microfluidic measurement are consistent with the theoretical model to a good extent statistically despite a number of limitations. Polystyrene spheres are investigated here, rather than the far more sophisticated bacterial cells. For instance, population growth, cell-cell communication, locomotion due to microvilli and flagella, specific rather non-specific DLVO interactions with the substrate, cell geometry and deformation, changing mechanical properties of cell surface due to environment, and the presence of nutrients and chemo-/mechano-taxis, etc. have inevitable influence on the experiments (Jacinta C. Conrad et al., 2011). Though a comprehensive theory is beyond the scope of this study, the present model serves as a reasonable first approximation shedding new light on colloidal filtration. In the classical picture, the DLVO potential with the 1min, 2min, and repulsive barrier, along with a stochastic description, is used to explain filtration efficiency, but does not include the influence of liquid flow. In fact, filtration efficiency and the rate of particle removal are shown to be dependent on the flow velocity, which is missing in the classical CFT. The commonly used column testing approach for measuring particle attachment coefficients does not consider the impact of varying flowrates on the results either. The relaxation time \( \tau \) which depends on the coupled ionic concentration and flowrate is also measured and incorporated into the present model in a phenomenological manner. For the long-term goal, the intrinsic biophysical parameters of live cells will be investigated and incorporated into the empirical equation. The theoretical model based on moment balance indicates the significance of inter-surface force and hydrodynamic interaction underlying the filtration mechanism.

It is worthwhile to list the experimental constraints that introduce possible artifacts in our measurements. When particles are injected into the channel prior to the liquid flow, the particle density has to be sufficiently low such that the inter-particle interactions are minimized and only a mono-layer, rather than stacked multi-layers, of particles are present on the substrate. The injection velocity of particle rich solution also plays an important role. Since our observation area is at the upstream front, high-speed injection causes the particles to spread out further from the inlet that might lead to a skewed distribution or nonuniform deposition on the substrate and thus inaccuracy in measurement and data scattering. Moreover, high injection pressure imparts high momentum that might unintentionally force more particles to overcome the repulsive
energy barrier reaching the primary energy well. The experimental procedures in this work were iterated a number of times to determine the optimal working parameters that minimized the undesirable effects.

5.6 Conclusion

A new microfluidic device was designed to investigate adhesion-detachment of colloidal polystyrene particles in an electrolyte in the presence of flow. A empirical expression for filtration efficiency was constructed based on the classical DLVO theory which comprises a double energy well. While the strong primary minimum adhesion prevents particles from escaping, the weak secondary minimum determines the dependency upon coupled flowrate and ionic concentration. When the solution salt concentration exceeds a critical threshold of 60 mM, flowrate no longer has an effect on the particle adhesion-detachment, because majority of the particles are trapped in the primary minimum. The new theoretical model based on moment balance was established. The new model sheds lights on the underlying physics of filtration. The results have significant impacts on in-situ or enhanced subsurface bioremediation, drinking water supplies, and water / wastewater treatments. The microfluidic device provides an economical and simple way to investigate live bacterial cells.
Chapter 6. New micro-fluidic experimental method on bacterial filtration efficiency and A Dynamic Tabor’s Parameter

In Chapter 5, the microfluidic tests were conducted on polystyrene particles of unique size and properties under varying flow rates and ionic concentrations. In this chapter, microfluidic tests are conducted on four different bacterial strains under varying flow rates and constant ionic concentration. The correlation between properties of individual strains and their filtration efficacies are found. We develop a dimensionless predictor of bacterial filtration efficiency coupling the effects of bacterial properties and flow condition, based on empirical considerations.

6.1 Introduction

Due to the complexity of bacterial filtration in a porous medium, direct measurement of bacterial filtration efficiency is impractical. Prediction of bacterial filtration efficiency from the traditional approach requires both the classical colloidal filtration theory (CFT) (Logan et al., 1995; Yao et al., 1971) and an empirical study, e.g. column test (Iwasaki et al., 1937; Y. Li et al., 2014; Tufenkji & Elimelech, 2004a; Tufenkji et al., 2004; X. Wang et al., 2012). The transport of bacteria in aqueous solution and collision between bacteria and single collector were solved theoretically using convection-diffusion equation (1-2) (Iwasaki et al., 1937; Rajagopalan & Tien, 1976). This was further improved by Yao et. All (Yao et al., 1971) by presenting an empirical formulation based on numerical results of convection-diffusion equation. This is so the called the classical colloidal filtration theory (CFT). The CFT equation was improved by Elimelech and Tufenkji by taking into account the impacts of electrostatic and van der Walls interactions (Elimelech, 1994; Tufenkji & Elimelech, 2004a). The total concentration of bacteria penetrating the porous system were measured experimentally from column test. Knowing the depth of column, grains size and porosity of medium, bacterial filtration efficiency of a single collector can be obtained.

The column test collaborating with CFT method gained its popularity in the last few decades in studying the impact of bacterial properties (Matthew W Becker et al., 2004; D. Fontes et al., 1991; Hendry et al., 1999; Ivanov, 2012; Kuznar & Elimelech, 2005; Rijnaarts et al., 1996b; Walker et al., 2004; Weiss et al., 1995), flow rate
(M. W. Becker, S. A. Collins, D. W. Metge, R. W. Harvey, & A. M. Shapiro, 2004; Camesano & Logan, 1998; Gannon et al., 1991; Hendry et al., 1999; Rijnaarts et al., 1996a; Tan, Gannon, Baveye, & Alexander, 1994) and ionic concentration (Bolster, Mills, Hornberger, & Herman, 2001; Deshpande & Shonnard, 2000; D. E. Fontes et al., 1991; Gannon et al., 1991; Hornberger et al., 1992; R. E. Martin, Bouwer, & Hanna, 1992) on filtration efficiency. However, the physics behind impact of each of those factors were usually not clearly explained. For example, bacterial softness and surface properties could affect its’ filtration behaviour via pathways like clogging between grains, attachment and detachment from collection surface (Y. Li et al., 2014). Contribution of each pathways is impractical to be identified. Column test results typically indicated a negative correlation between flow velocity and filtration efficiency (Bergendahl & Grasso, 2000; Hendry et al., 1999; X. Li et al., 2005; Shen et al., 2010; Tong & Johnson, 2006; Torkzaban et al., 2015; Torkzaban et al., 2007). There could be, but not limited to, the following two explanations: (i) higher flow rate break through the clogging caused by colloidal/bacterial aggregation (Bergendahl & Grasso, 2000; X. Li et al., 2005; Shen et al., 2010; Torkzaban et al., 2015; Torkzaban et al., 2007) (ii) colloid/bacteria adhered to the collector surface are detached by the hydrodynamic drag force (Tong & Johnson, 2006). The column test does not provide detailed information on which explanation is more likely or exclusively correct.

The limitations of the column test above are due to the complexity of porous medium filtration. In this study, we are presenting a new experimental method to tackle issues we discussed above. A microfluidic channel was assembled by glass slide and the polydimethylsiloxane (PDMS) as a micro filtration system. In the new microfluidic test, the only trapping mechanism is the surface adhesion between glass slide surface and bacteria. The potential interruption of other trapping mechanisms, for example straining and clogging, are excluded in this test method. Due to the transparency of the new experimental set up, detachment of bacteria was observed directly using optical microscope. A software with user interface was developed to process the test images and count the number of bacteria adhered to the glass surface. Micro channel can be dissembled after test and is reusable following the cleaning procedure. Another significant advantage of the new method is time efficiency. The sample volume required by each test is reduced tremendously thus the time consumed for each test is from 30 seconds to 10 minutes depending on flow rate, comparing to test time of hours to days need for column test (Y. Li et al., 2014). Four different bacterial strains
with different microscopic properties, measured by atomic force microscopy (AFM) were tested under different flow velocities. Attachment efficiencies were measured directly from the microfluidic tests and compared with the results obtained by the conventional column test. We studied the impact of micro properties of individual bacteria in terms of Tabor’s parameter on its’ filtration efficiency. Effect of flow velocity on filtration efficiency was also investigated. At last we developed a dynamic Tabor’s parameter as an indicator of bacterial filtration efficiency which couples the effects of mechanical properties of bacteria and the flow conditions.

6.2 Experiment and Method

6.2.1 Experiment

The experimental setup has three main sections, syringe pump (model NE-300, New Era Pump System Inc., Farmingdale, NY), microfluidic channel and microscope integrated with PC, as shown in the Figure 5.1 (b). Working with 1mL Luer Lock Tip Syringe (by EXELINT) with inner diameter of 4.65 mm, the pump is able to achieve the lowest flow rate of 0.714 μL/h. To achieve the range of groundwater flow velocity, 0.1 to 10 meter per day, dimension of the channel is designed to be 70 μm in height and 3000 μm in width. Before the test, the micro channel is placed on the platform of an inverted optical microscope (model GX71, Olympus Inc., Waltham, MA). An objective lens with magnification of 100× was adopted. The tests were monitored in real time via the screen shown in the Figure 5.1 (b) and micrographs were taken before and after the onset of flow.

6.2.2 Microfluidic Channel Preparation

The channel mold is prepared in a circular clear quartz petri dish. A Scotch tape™ (3M) is firstly cut to desired dimension and measured. Then it is placed on the surface of petri dish. The tape forms the chamber void. PDMS is poured (Sylgard 184; Dow Corning) onto the mold and vacuum it. When there are no air bubbles in the PDMS we put the mode into the oven and cure it at 69°C for 2 hours. After cured, two inlets holes and one outlet hole were drilled into top PDMS slab using a punch. The second inlet for loading bacterial solution is made at the mid span. The other two holes are made at each end of the channel as shown in the Figure 6.1.
The glass slide is sonicated and PDMS is cured in DI water for 1 hour, followed by treating the resin in isopropyl alcohol and another sonication for 1 hour. The glass slide and PDMS are dried using high pressure Nitrogen spray. Then we bond the glass slide and PDMS by gently pressing from the center outwards to squeeze all the air bubbles. Lastly the pre-bonded channel is placed on a hot plate of 85°C for 2 hours, while subjected to a gentle press on top of the channel. This bonding method does not provide adhesion as strong as the oxygen plasma method provides. But the adhesion is strong enough to meet the pressure requirement of the experiment in this study. Another advantage of this bonding method is that it allows the reuse of the channel. This is significantly cost effective and time efficient. After each experiment, the micro channel is dissembled by peeling glass slide and PDMS apart. Glass slide and PDMS are cleaned by DI water and followed by resin in isopropyl alcohol for future reuse.

6.2.3 Other materials

The background electrolyte solution used in this study is potassium chloride (KCl) with ionic concentration of 3 mM. This value is in the range of groundwater ionic concentration and it is identical to the value adopted in our previous column test and AFM test. In this study, we used the same four bacterial strains as previous column tests.
for comparison purpose as follows: (i) *Shewanella oneidensis* MR-1, (ii) *Bacillus cereus*, (iii) *Raoultella ornithinolytica* and (iv) *Aeromonas punctate*. The background information and mechanical properties of the four strains are summarized in the Table 1.1. *Shewanella oneidensis* MR-1, strains SH1, was grown anaerobically in Luria-Bertani (LB, Sigma-Aldrich, Inc., St. Loues, MO) medium (25 g/L) with 10mM sodium fumarate (1.6 g/L) as electron acceptor and 10 mM sodium lactate as electron donor. Strain SH1 was cultured at 30 °C in a glove box with an atmosphere of 5% hydrogen/nitrogen balance. *Bacillus cereus*, *Raoultella ornithinolytica* and *Aeromonas punctate*, corresponding to strain H, A, Q, are aerobic and were cultured in 25 g/L Luria-Bertani (LB) medium in 50 mL tube at 37˚C. To eliminate the impact of growth stages on bacterial adhesion properties, all bacteria were harvested from the stationary stage (16~18 hours growth) by centrifugation. Then centrifuged bacteria were resuspended in 30 mL of background solution for experimental usage.

### 6.2.4 Experimental Procedure

We injected background solution (KCl) from the inlet 1, shown in the Figure 6.1, and fill the channel with background solution. Then we injected the bacterial solution from the inlet 2. The volume of bacterial solution injected was carefully controlled such that bacterial solution only filled the mid-portion of the channel and form a boundary between bacterial solution and background solution at upstream. Thus, when we focus on the region close to the boundary, no bacteria from the upstream region would enter the focus region and disturb the counting of bacteria.

A wait time of 1-10 minutes for bacteria to settle was allowed. Some bacteria would adhere to the glass surface and some remain suspended in the solution. The test for the desired flow velocity was started following the capture of an initial picture. Flow velocities tested in this study were set to meet two requirements: (i) cover the flow velocities of 0.015, 0.06, 0.09 cm/s and 0.15 cm/s, used in column tests, (ii) cover the lower end of the groundwater flow velocity 1.5 - 15 μm/s. Only strain A and Q were tested using the lowest flow velocity of 1.5 μm/s. We took snapshots with frequencies that varied from 10 seconds to 2 minutes depending on the flow velocity. High flow velocity requires small time step to increase resolution. When no bacteria move in the focused region we considered the system was stable, all bacteria retained were successfully attached on the glass slide under the tested flow velocity. For the lowest flow velocity of 0.00015 cm/s, it usually took 10-20 minutes for the system to stabilize.
For the highest flow velocity of 0.15 cm/s, the whole process usually took 1 minute. Each experimental condition was run in triplicate. After each test, the micro-chamber is dissembled and cleaned as described in the microfluidic channel preparation section.
6.2.5 Image Processing

![Image of raw image, binary image, binary image with noise elimination, bacterial aggregation with one bacterium, and bacterial aggregation with 2, 3, 5, and 7 bacteria.]

Figure 6.2. Solution for bacterial counting. Here the raw image of strain A after settling on the channel substrate is shown in (a). As part of image processing, we determine a critical threshold grayscale level (0.49 in this case) and utilize this value to convert raw image to binary image, shown in (b). Then we determined a range of pixel number (40 to 1000 in this case) to cover all white dots we considered as bacteria. Utilizing this range, we excluded all noise and derived a binary image ready for counting, shown in (c). Part 2 is bacterial counting. In this part, we first determine the number of pixels that a single bacterium occupies (245 in this case). Thus, single bacteria were extracted and counted, as shown in (d). Second, we determined the number of pixels for bacterial aggregation (130 in this example). Using the same method, bacterial aggregation with 2, 3 … bacteria can be extracted, as shown in (e). In this example, only aggregation with 2, 3 and 7 bacteria were detected.
6.2.5.1 Bacterial Counting

We developed a program to count the number of bacteria with user interface (UI) in MATLAB. The software consists of two parts, image processing and bacterial counting. A typical raw image is shown in Figure 6.2(a). We adjusted the microscope, focusing on the bacteria that contacted the chamber substrate. These bacteria are thought to include ones in both primary minima and secondary minima. In Figure 6.2(a), these bacteria are indicated by the dots with high light intensity. The dots with dark color indicate bacteria that suspended about 1~10 microns away from the substrate and they are not in either primary minima or secondary minima thus not considered in our counting.

During image processing section, we converted the raw image into a binary image. In a perfect conversion, all bacteria contacting the glass substrate would be converted into white colored with black background. Due to non-uniform distribution of illumination, it is hard to obtain a perfect conversion. To achieve a good conversion, we developed a two-step solution. First, we find the best threshold value of grayscale level by trying multiple values. After this step, we obtain a binary image with noise, as shown in the Figure 6.2(b). In the second step, we find the critical range of number of pixels to cover all bacteria beyond which the number is either too small or too large to indicate a single bacteria or bacterial aggregation. After the second step, we obtained the binary image ready for bacterial counting, as shown in the Figure 6.2(c). The whole process requires carefully manual investigation in order to obtain the critical threshold grayscale level and a range of pixel numbers indicating the bacteria. Take the image in the Figure 6.2(a) for instance, the critical threshold grayscale level is 0.499 and range of number of pixels is from 40 to 1000.

In Figure 6.2(c), each dot represents an individual bacterial aggregation that occupies different number of bacteria. Thus, in the bacterial counting section, we first extract the dot representing single bacteria by setting the number of pixels a single bacterium occupies, as indicated in the Figure 6.2(d). Second, we extract the dot with increasing number of bacteria aggregation, as 2, 3 etc. as shown in Figure 6.2(e). In this process, the number of pixels increment for bacterial aggregation increase was determined ahead of time. Therefore, the total number of bacteria in the image is derived as
\[ N = \sum_{i=1}^{M} i \cdot n_i \]  

where \( N \) is the total number of bacteria in the original image, \( i \) represents bacterial aggregations containing \( i \) number of bacteria, \( M \) represents the bacterial aggregation containing the largest number of bacteria and \( n_i \) is the number of bacterial aggregations with \( i \) number of bacteria.

6.2.5.2 Analysis of the Orientation of Bacteria

The magnitude of the hydrodynamic interaction exerted on individual adhered bacteria depends on the orientation angle between the bacteria and the flow direction, indicated by \( \theta \) in the Figure 6.3. High hydrodynamic drag force is expected when bacteria is perpendicular to the flow direction, i.e. \( \theta=90 \) degrees. Low magnitude of hydrodynamic drag force is expected when bacteria are aligned with the direction of the flow. Therefore, to achieve better understanding of fate of bacterial detachment, it is helpful to study the orientation distribution of the bacteria in the presence of flow.

![Figure 6.3. Schematic of bacterial orientation in the presence of flow. The inclination angle \( \theta \) is angle between direction of bacterial long axis and flow direction.](image)

An algorithm was developed using Python 3.0, shown in Appendix 2, to draw backbone of individual bacteria Figure 6.4 (a) shows the original micrograph of strain A initially adhered on the glass slide and Figure 6.4 (b) shows the corresponding binary image of bacterial backbones. The coordinates of the pixels at the end points of each backbone are obtained. Thus, the orientation angles of bacteria can be derived through an inverse trigonometric relationship between position of two ends of bacteria and its’ orientation.
Figure 6.4. (a) Initial state of strain A adhered on the glass slide before flow. (b) Binary image indicating the backbone of bacteria. The arrow in the image indicates the direction of flow.

This study was conducted on strain A, H and Q. The algorithm was applied to the first micrograph before the test and the last micrograph after the test. The results between the two were compared to study the effect of flow on bacterial orientation. Varying flow velocities of $1.5 \times 10^{-6}$~$1.5 \times 10^{-3}$ m/s were tested. Each test has a minimum of 3 repeats.

6.2.6 Modified Tabor’s Parameter

A strong correlation between bacterial micro properties, measured by atomic force microscopy (AFM), and its macro filtration efficiency measured by column test was concluded (Y. Li et al., 2014; X. Wang et al., 2012). The micro-macro properties were bridged by a modified Tabor’s parameter. Derivation of the modified Tabor’s parameter is briefly introduced in the following paragraph.

A 1 mL suspension of bacteria culture in 3 mL KCl solution was pipetted onto a gelatin-treated, cleaved mica disk (Sigma G-6144) for AFM indentation. The mechanical response was derived by loading and unloading of the AFM tip. From the mechanical response, the following quantities were derived: $b$ is the width of the ellipsoidal bacteria, $l$ is the thickness of the cellular surface substances (CSS), $E$ is the bacterial elastic modulus, $U_{ad}$ is the total adhesion energy needed to detach the AFM tip from bacteria, $R_{AFM}$ is the AFM tip radius and $\nu$ is the Poisson’s ratio. More detailed measurement procedure could be found from a previous publication (Y. Li et al., 2014). With the attempt to derive a universal dimensionless parameter to reflect the bacteria-surface interaction quantities as well as the bacterial dimension and mechanical
properties, the modified Tabor parameter (Xin Wang, 2013), \( \mu \), was defined by combining the measured quantities listed above as

\[
\mu = \left[ \frac{2b}{l^3 E^2} \left( \frac{U_{ad}}{\pi R_{AFM}} \right)^2 \left( 1 - \nu^2 \right)^2 \right]^{1/3} \tag{6-2}
\]

All measured quantities and modified Tabor’s parameter \( \mu \) of four bacterial strains are listed in the Table 1.1. A more detailed description of the basis and development of the modified Tabor’s Parameter can be found in reference (Xin Wang, 2013). Bacteria with a larger Tabor parameter indicates it is larger, more compliant and stickier. Therefore, once bacteria with large \( \mu \) adheres to a surface we expect it is more difficult to be detached. This hypothesis was supported by the column test which measure the filtration efficiency of individual bacteria. The results show a significant correlation between the Tabor and the filtration efficiency (Y. Li et al., 2014) as shown in Figure 6.5. The four strains investigated in this study are ranked as A< SH1< Q< H in terms of Tabor Parameter.

In this study, we examined the correlation of modified Tabor parameter and bacterial filtration efficiency measured by microfluidic test which will be discussed in the next section.

We further developed a modified Tabor parameter by including the interaction introduced by flow velocity. The new Tabor parameter is reflecting a coupled effect of bacterial micro properties and fluid interaction.
Figure 6.5. Correlation between Filtration efficiency derived by microfluidic test (MF test) and Tabor’s parameter of four bacterial strains under different flow.
6.2.7 A Dimensionless Hydrodynamic Factor

Although the modified Tabor parameter captured the correlation between bacterial micro properties and macro filtration behavior, it does not reflect the interaction of the hydrodynamic force and adhesion exerted on bacteria. This is because the quantities in the Tabor parameter are independent of the flow velocity. To take into account the effects of the flow condition, we developed a dimensionless hydrodynamic factor based on the Reynolds number. This factor includes properties of fluid, e.g., density and dynamic viscosity, and flow velocity. The flow velocity affects via hydrodynamic interaction exerted on bacteria thus bacterial dimension is also included in this factor. The hydrodynamic factor is expressed as follows:

$$\beta_{\text{hydro}} = (C \times \frac{LV}{\tau})^m = (C \times \text{Re})^m$$  \hspace{1cm} (6-3)$$

where $L$ is the length of long axis of bacteria, $\tau$ is dynamic viscosity of water, $V$ is average flow velocity in the channel, $\rho$ is water density, $\text{Re}$ is the Reynolds number, $C$ and $m$ are adjustable parameters. Collaborating with the modified Tabor’s parameter discussed in the previous section, a new parameter for bacterial filtration efficiency is defined as follows:

$$\mu_c = \beta_{\text{hydro}} \times \mu$$  \hspace{1cm} (6-4)$$

This new parameter takes into account both colloidal/bacterial properties and fluidic conditions. Parameters $C$ and $m$ are determined by fitting $\mu_c$ and filtration efficiency to Pearson correlation observed in experiments (Y. Li et al., 2014).

6.3 Results and Discussion

6.3.1 Attachment Efficiency of Different Flow Velocities

Figure 6.4 shows the attachment efficiency derived from the new microfluidic test under different flow velocities. The lowest flow velocity of 1.5 $\mu$m/s was conducted on strain A and Q. Only 2% to 5% of bacteria were detached under low velocity. When flow velocity was increased to 0.015 cm/s, detachment ratio increased to 40-60% depending on the bacterial species. When the flow velocity was increased from 0.015 to 0.15 cm/s, detachment ratio was increased 5-25% depending on the bacterial species. A special case is strain H which is usually sticky. Detachment ratio of strain H is within 10% under all tested flow velocities. This observation is consistent with previous
column test results (Y. Li et al., 2014) and surface force measurements (Xin Wang, 2013).

Figure 6.6. Impact of flow velocity on filtration efficiency for four bacterial strains. Tested flow velocity range covered the typical flow velocity of groundwater and flow velocity adopted in the previous column test.
Effect of flow velocity on bacterial filtration is due to interaction between hydrodynamic moment and adhesive moment exerted on the bacteria. Adhesive moment that bacteria experience under given ionic concentration is not constant. There are two reasons. First the bacteria in the background solution is charged differently and this results in a probabilistic distribution of the zeta potential for all bacteria. Bacteria with low zeta potential could achieve strong adhesion. Second, the orientation of the bacteria results in different adhesive moment to resist detachment interaction, hydrodynamic moment. The more aligned with the flow direction the bacteria are, the harder it will be to detach them. The difference in the behavior of different bacterial strains is due to the different micro scale properties. This will be discussed in the next section.

6.3.2 Correlation of microfluidic test results and AFM measurement

All measured quantities from AFM test contributed to build the modified Tabor parameter defined in Eq(6-2). In terms of Tabor parameter, the four strains are ranked as A< SH1 <Q <H. The modified Tabor parameter governs the compliant profile, contact area and adhesion interaction once bacteria adhere to micro-channel substrate. Bacteria with large Tabor parameter deform drastically, making it more geometrically streamlined and thus experiencing less hydrodynamic interaction, for instance the strain H. Pearson correlation between the Tabor parameter and the filtration efficiency of bacteria measured by column test was observed (Y. Li et al., 2014), as indicated in Figure 6.5. Comparison of this results with the results of the microfluidic test shows agreement between the two. The microfluidic test is able to capture the impact of bacterial micro properties on its filtration behavior. For instance, strain H has high adhesion energy and the thin CSS layer among the four tested strains. These characteristics could help strain H to achieve strong adhesion in the primary minimum and thus to resist detachment from hydrodynamic interaction.
Figure 6.7. Correlation between filtration efficiency and Tabor’s parameter and hydrodynamic factor. Parameters in $\beta_{hydro}$ were derived as, $C = 1.07 \times 10^{-11}$ and $m=1/3$. Tabor’s parameter $\mu$ was derived from AFM analysis. All strains were in stationary stage. Only strain A and Q were tested under the lowest flow velocity of 0.00015 cm/s. Attachment efficiency derived by column test is also presented in the figure and marked as red star. Column test was tested under 0.015 cm/s and MF test under same flow velocity is heighted. Agreement between the two was indicated.
Microfluidic tests were conducted at flow velocities of 0.0015, 0.015, 0.06, 0.09 and 0.15 cm/s. As discussed in the previous section, filtration efficiency tends to decrease as flow velocity increases. This is because hydrodynamic interaction increases with the flow velocity. Note that the flow velocity affects different bacteria differently. A coupled impact of bacterial type, indicated by Tabor’s parameter, and flow velocity, indicated by $\beta_{\text{hydro}}$ on filtration efficiency is plotted in the Figure 6.7. Compared to other strains, strain H is most compliant on glass surface and most adhesive. These properties resulted in strong adhesion of strain H such that hydrodynamic force has no effect on it. Detachment of other strains is subject to flow velocity. Impact of flow velocity on the attachment of other bacterial strains decrease in the order of their Tabor’s parameter. Strain A and Q were tested at the lowest flow velocity of 0.00015 cm/s. The filtration efficiency of the two bacteria under this flow velocity are around 0.96-0.99. Adhesion of bacteria overcomes hydrodynamic interaction thus detachment rarely occur.

**6.3.3 Dynamic Tabor’s Parameter**

The coupled impact of bacterial type and flow velocity on filtration behavior can be presented quantitatively using the new Tabor’s parameter shown in the Eq(6-4). A correlation between the new Tabor’s parameter and attachment efficiency is shown in the Figure 6.8 (Pearson correlation, $r_p = 0.539$, $p = 0.051$). By fitting the experimental results to the Pearson correlation, the parameters $C$ and $m$ in Eq(6-3) can be determined as, $C = 1.07 \times 10^{-2}$ and $m=-1/3$. The new relationship connects the macroscopic behavior of bacteria to its microscopcic properties, along with the flow conditions. It not only provides a powerful tool on predicting different bacterial filtration efficiency but also shed new lights on understanding the physics mechanisms behind bacterial filtration.
Figure 6.8. Pearson correlation between attachment efficiency ($\alpha$) derived by microfluidic test (MF test) and new predictor, $\beta_{\text{hydro}} \cdot \mu$. $\mu$ is modified Tabor’s parameter which was derived from AFM analysis. Error bar represent the standard deviation from at least triplicated MF test. All strains were in stationary stage. Only strain A and Q were tested under the lowest flow velocity of 0.00015 cm/s.

### 6.3.4 Distribution of Bacterial Orientation

In this section, we look into the impact of bacterial orientation on their detachment behavior. Tests were conducted on three different strains, strain A, H and Q. The experimental procedure was illustrated in detail in the Section 5.2.4. In brief, using the syringe pump, the bacterial solution was slowly injected into the micro-channel filled with background solution via inlet 2. Once desired amount of bacteria (roughly fill one third of channel volume) was injected, the syringe pump was terminated immediately and bacteria settled down on the glass substrate due to gravity. In such a way, we aimed at eliminating the effect of injecting flow velocity on bacterial settlement on the glass surface. A micrograph was taken at initial state of bacterial settlement. For instance, Figure 6.4 (a) shows initial state of strain A, settling on the
glass slide. This was followed by a flushing through of background solution under desired flow velocity of $1.5 \times 10^{-6} \sim 1.5 \times 10^{-3}$ m/s until the system reaches the equilibrium. A micrograph was taken at the final equilibrium state. By processing the micrographs, orientation of bacterium with respect to the flow direction was measured both at initial state of settlement and final equilibrium state in the presence of flow. Histograms of orientation angles are shown in Figure 6.9 for all three strains and varying flow velocities.
Strain H

1.5\times 10^{-5} \text{ m/s}

1.5\times 10^{-4} \text{ m/s}

6\times 10^{-4} \text{ m/s}

9\times 10^{-4} \text{ m/s}

1.5\times 10^{-3} \text{ m/s}

(a)

Probability of Orientated Angle

Orientated Angle, \theta (Degree)
Strain Q

1.5 × 10^{-6} m/s

1.5 × 10^{-5} m/s

1.5 × 10^{-4} m/s

6 × 10^{-4} m/s

9 × 10^{-4} m/s

1.5 × 10^{-3} m/s

Probability of Orientated Angle

Orientated Angle, θ (Degree)
Figure 6.9. Histogram of bacterial orientation angle before and after the flow, indicated by grey bars and red bars respectively. (a). Strain H (b). Strain Q (c). Strain A.
First, we investigate the pattern of histogram at the initial state. Strain Q and strain A showed no apparent preference on the orientation angle of their settlement on glass slide. This is indicated by the grey bars in Figure 6.9(b) and (c). Strain H showed a slight half-bell pattern, indicated by grey bars in Figure 6.9(a). This implies that, during settlement, strain Q and A tend to orientate randomly, yet strain H tends to orient itself in alignment with the flow direction of initial injection. We suspect that even with careful control of initial bacterium settlement as we described in previous paragraph, initial injection of bacterium introduced velocity and this velocity affected orientation of strain H while they were settling on the substrate. Yet effect of initial velocity on orientation of the other two strains in their settlement are negligible.

Next, we investigate the pattern of histogram when the system reaches the equilibrium state after flushing through background solution, indicated by the red bars in Figure 6.9. In order to better explore the effect of flow on detachment of different orientation angles, the detachment ratio of bacteria of different orientation angle was plotted as shown in the Figure 6.10. This detachment ratio is defined as the fraction of bacteria that were detached under a specific orientation angle. For example, in Figure 6.10(b) of strain Q, with the flow rate of $9 \times 10^{-4}$ m/s, only 2% of bacteria with orientation angle of $<5^\circ$ were detached, and 23% of bacterium with orientation angle of $45^\circ$ were detached. In case of negative detachment ratio, as shown in Figure 6.10 (a), it means the number of bacterium with this specific orientation angle increased after the test. This could be caused by the following two reasons: (i), some of bacterium were oblique with flow at initial state and later on the bacterium were driven to be more aligned with flow by hydrodynamic interaction; (ii). some bacterium from upstream of channel were flushed into the focused region, attaching on the glass slide and parallel with flow. Since the impact of bacterium from the upstream was minimized in the current experimental setup, we believe the negative detachment ratio was mainly due to reason (i). This was frequently observed in the test of strain H, as indicated by Figure 6.10(a). Therefore, we believe that the bacterium of strain H of high orientation angle was re-oriented by hydrodynamic interaction and parallel with flow, instead of being full detached. For strain A and Q, we believe that re-orientation and fully detachment both occurred for bacteria of high orientation angle, yet we did not see a strong correlation between detachment ratio and initial orientation angle of bacteria.
Strain H

Detachment Percentage of Different Orientation Angle, %

- Orientation Angle, θ (degree)

(a)
Strain Q

Detachment Percentage of Different Orientation Angle, %

Orientation Angle, θ (degree)

- $1.5 \times 10^{-6}$ m/s
- $1.5 \times 10^{-5}$ m/s
- $6 \times 10^{-4}$ m/s
- $9 \times 10^{-4}$ m/s
- $1.5 \times 10^{-3}$ m/s
(b)

Strain A

Detachment Percentage of Different Orientation Angle, %

1.5×10^{-6} m/s

1.5×10^{-4} m/s

6×10^{-4} m/s

9×10^{-4} m/s

1.5×10^{-3} m/s

Orientation Angle, θ (degree)

(c)
Figure 6.10. Detachment fraction of bacterium with different orientation angle of $\theta$.

6.4 Conclusion

First, a good agreement between results of microfluidic and column tests was observed, shown in the Figure 6.5. We believe the new microfluidic test method has a great potential to take place of column test when experiment meets the following two conditions: (i) measuring filtration efficiency when particle size is much smaller than void size (ratio of diameter $< 1:50$) (ii) surface absorption is the main filtration mechanism. The microfluidic test provides a direct way to measure filtration efficiency and is more time efficient compared to the column test.

Second, the microfluidic test captured the coupled effect of bacterial properties (Tabor’s parameter) and flow condition on filtration efficiency. This motivated us to integrate the hydrodynamic interaction into the existing Tabor’s parameter and derive a dynamic Tabor’s parameter, a combination of bacterial properties and flow condition. Figure 6.8 showed a strong correlation between bacterial filtration efficiency and the dynamic Tabor’s parameter. A special case was strain H. Filtration efficiency of strain H is not sensitive to flow velocity because its surface adhesion is so strong that it dominated bacterial detachment under all flow velocities. Meanwhile, strain A, Q and SH2 exhibited a low surface adhesion and high elastic modulus. Their detachment ratio was subject to their micro-properties and flow rate.

Since hydrodynamic force exerted on a bacterium is a function of the orientation. We investigated the effects of the orientation angle of the bacteria on their detachment under flow. We found that detachment of strain Q and strain A randomly occurred in a span of inclination angles of $0^\circ$ to $90^\circ$. For strain H, the bacterial detachment rarely occurred due to strong surface adhesion. However, bacteria of strain H would orientate themselves and align with flow direction in the presence of hydrodynamic interaction. This further enhanced the attachment of the strain H on the collector.
Chapter 7. A New Filtration Model on Bacterial Filtration in Porous Medium

7.1 Introduction

In both water treatment industry and natural systems, filtration is an important process to eliminate toxic particles or microbes from water. In contrast to chemical methods and other disinfection methods with high costs and toxic byproducts, filtration is environmentally friendly and cost-efficient. Study of filtration helps understand how microbes and particles transport underground. For example, how far should a water well be placed away from polluted water source can be discerned from such studies.

As the most widely used indicator of filtration efficiency, the single collector attachment efficiency $\alpha$ is defined as the ratio of number of colloids/bacteria that attach on a single collector to the number of colloids/bacteria that collide with it (Tufenkji & Elimelech, 2004a; Yao et al., 1971). The filtration process involves two separate steps: the transportation of suspended colloids/bacteria to the collector surface and successful attachment of colloids/bacteria that collide with the collector surface. Thus, calculation of $\alpha$ involves two aspects.

The first aspect is to calculate the number of colloids/bacteria collide with a single collector. Traditionally this is done by solving the general equation, Eq (1-2), describing the temporal and spatial variation of particle concentration. This equation is derived from a mass balance of particles about an elemental volume of suspension. A numerical solution provides the particle concentration distribution on a collector and the number of particles that collide with the collector can be thus found by integrating the concentration with respect to the entire collector surface. In 1971, Yao addressed three mechanisms dominating the transportation process, as diffusion, interception and sedimentation (Yao et al., 1971). By fitting the numerical results obtained from solving the transport equation (1-7), Yao derived an empirical equation to determine the single collector collision efficiency $\eta$ with three terms, representing the three mechanisms respectively.

The second aspect is to derive the number of colloids/bacteria captured by a single collector. Performance of packed bed were evaluated by column tests. Relation between bed depth and number of colloid/microbe filtered were developed by Iwasaki in 1937 (Iwasaki et al., 1937). Yao further developed this method and related the performance of a sand bed to the filtration efficiency of a single collector as,
\[
\ln \frac{C}{C_0} = -\frac{3}{2} (1 - \varepsilon) \alpha \eta \frac{L}{d}
\]

where \(C\) and \(C_0\) are the effluent and influent colloidal concentration of a sand bed, \(L\) is bed depth, \(d\) is collector diameter and \(\varepsilon\) is bed porosity. \(C\) and \(C_0\) are measured by laboratory experiment, or column test. The single collector attachment efficiency \(\alpha\) is thus derived. The method that combines these two aspects is known as the classical colloidal filtration theory (CFT).

The CFT theory requires a combined approach where first the number of collisions is computed and second the number of attached particles is determined experimentally. In the last few decades, the CFT theory has been widely used to evaluate colloidal/bacterial transportation in porous medium. The improvements made on the CFT theory were only focused on the first part. In 1977 Rajagopalan and Tien (Rajagopalan & Tien, 1976) developed their own model using trajectory analysis to calculate the single collector efficiency \(\eta\). Elimelech and Tufenkji (Tufenkji & Elimelech, 2004a) improved the correlation equation of \(\eta\) by including the van der Waals force in 2004. Information provided by \(\eta\) is very limited because most of information are mined in the packed-column experiments. For example, in investigation of bacterial filtration behavior, bacterial motility (Camesano & Logan, 1998; Hendry et al., 1999), stiffness, geometry, surface substance (M. W. Becker et al., 2004) and biological factors (Syngouna & Chrysikopoulos, 2011) can be accessed only by column test. Effects of flow rate and ionic concentration on filtration efficiency are mostly embedded in the column test. Hydrodynamic interaction is considered as a dominant mechanism of colloidal/bacterial detachment after collisions occur (Bergendahl & Grasso, 2000; Shen et al., 2010; Torkzaban et al., 2007). The DLVO interaction has a very short effective range, within 100 nm according to our calculation. Thus, effect of electrostatic double layer on suspended particles in bulk is usually negligible. However, DLVO interaction does affect the colloids/bacteria that already contact the collector. Although these effects are accessible by the experiments, the mechanisms behind them are not clear.

In this study, we present a conceptual model for bacterial attachment/detachment after collision based on comparing the adhesive moment and hydrodynamic moment. Multiple theoretical models have been built based on the force/moment balance between hydrodynamic interaction and surface adhesive interaction (Bergendahl & Grasso, 2000; Scott A. Bradford et al., 2009; Shen et al.,
The velocity field in packed beds of porous media has been represented mainly by three models: sphere-in-cell, capillary tube and constricted tube (Bergendahl & Grasso, 2000). Sphere-in-cell model was applied in this study. Hydrodynamic moment of a cylindrical particle with varying orientation angles against flow were derived using computational fluid dynamics. A cylinder contact model based on Maugis analysis (K. Johnson & J. Greenwood, 2008) and Derjaguin, Landau and Verwey, Overbeek (DLVO) theory was adopted in calculating adhesive moment. Size, geometry and elastic modulus of bacteria are considered in this model. Previous AFM (atomic force microscopy) tests on bacteria showed strong correlation between bacterial properties and filtration efficiency (Y. Li et al., 2014; X. Wang et al., 2012). To verify the new model, we conducted well controlled column tests on two different bacterial types, Aeromonas punctate (strain Q) and Raoultella ornithinolytica (strain A), under different flow velocities and different ionic concentrations with constant temperature and bacterial concentration. This model, replacing the conventional column test, collaborating with the CFT theory provides a theoretical method to predict filtration efficiency of bacteria. This model provides a great potential to avoid experiments and serve as a platform to study the effects of bacterial micro-properties on filtration efficiency.

7.2 Material and Method

7.2.1 Experiment

7.2.1.1 Bacterial preparation

Two bacteria strains to be investigated have relevance related to environment and human health (Y. Li et al., 2014). Strain Q: *Aeromonas punctate*, Gram-negative bacteria are reported to be associated with human diseases including gastroenteritis, cellulitis and diarrheal. Strain A: *Raoultella ornithinolytica*, Gram-negative bacteria are a major cause of histamine fish poisoning (X. Wang et al., 2012). Both strains are aerobic and were grown aerobically at 37 °C in 25 g/L Luria-Bertani (LB) medium (Sigam-Aldrich, Inc., St. Louis, MO). Here we only summarize the properties under ionic concentration of 3 mM. To eliminate the potential impact of the growth stage on bacterial micro adhesion properties (Y. Li et al., 2014), all sample cells are in stationary growth phase, namely 16-18 hours growth.
Figure 7.1. Schematic of Column test. The column is packed with sand grains with diameter of 289 μm. Bacterial suspension was pumped through the column followed by background solution. Bacterial concentration at the column outlet is monitored by spectrophotometer.
7.2.1.2 Experimental Procedure

Schematic of experimental setup is shown in the Figure 7.1. Ultrapure silica sand GRANUSIL 4095 (UNIMIN Corp., LeSueur, MN) with a nominal diameter of 289 µm was utilized as collector in the column testing. The sand was cleaned with 1 mM NaOH for 24 hours, rinsed with DI water, dried in an oven at 103 °C for 24 hours, followed by drying in a 550 °C oven for 1 hour before use. A suspension of bacteria was pumped through a 60 cc sterile syringe with inner diameter of 2.67 cm, packed with clean silica sand to a height of 10 cm (Y. Li et al., 2014). A column packing porosity of 0.4 was determined by applying standard gravimetric methods (Y. Li et al., 2014). 20 pore volumes of DI water followed by 10 pore volumes of the background electrolyte solution (3 mM Potassium Chloride solution) were pumped through the syringe at a certain flow rate, ranging from 5~50 mL/min (filtration velocity 0.015 cm/s ~ 0.15 cm/s), before each test to make sure the packed column was equilibrated. A suspension of bacteria in the same background electrolyte solution was pumped for 3-4 pore volumes (PV, 1PV=24 mL), followed by bacteria-free background electrolyte solution (about 3 pore volumes) at the same rate as velocity used in previous step. To make sure the bacteria concentration to be constant during test, a miniature magnetic stir bar was applied in the bacteria solution tank and the bacteria concentration at the inlet of column was measured at the beginning and the end of duration of pumping bacteria solution. The bacteria concentration at the outlet of column was monitored continuously using optical density measurements at 500 nm with a UV-vis spectrophotometer (model UV Mini 1240 Shimadzu, Kyoto, Japan) and a 1 cm flow through cell.

7.2.2 Theory

7.2.2.1 Geometry

Figure 7.2 describes the geometry of bacterial suspension flow through the packed sand column from bottom to top. The bacterial suspension flow through the packed sand, some bacterium collided with a sand grain and adhere to the sand surface. The compact arrangement of sand grains in our column test are assumed to be cubic packing. The porosity of cubic packing is 0.476 by calculating ratio of void volume to bulk volume and this is reasonably close to the porosity value of 0.4 measured in our experiment. As geometry is axisymmetric in the flow direction, indicated by blue arrows in the Figure 7.2, the attachment position on the collector can be determined by
one parameter $\varphi$. In Happel’s sphere-in-cell model (Happel, 1958; Shen et al., 2010), the hydrodynamic stress distribution on the collector only depends on $\varphi$. Bacteria, adhering on sand surface, has orientation angle of $\theta$, indicating the angle bacterial orientation with respect to local flow direction, shown in Figure 7.2. The hydrodynamic interaction on an adhering bacterium depends on $\varphi$ and $\theta$. 
Figure 7.2 Bacterial suspension flow through packed sand medium. From bottom to up, the figure indicates: bacterial suspension flow through...
cubic packing of sand grains; bacterial suspension flow around a single sand grain; bacterium collided with collector and adhered on the collector surface. The packing pattern of sand grains in this study is assumed to be cubic pattern. Because the porosity of cubic packing is close to the porosity measured in the column test. The arrows indicate the upper flow direction. Location of a bacteria on collector is determined by \( \phi \). Due to axisymmetric, hydrodynamic stress is constant in the grey region, defined by \( 2\pi \cdot \sin(\phi) \cdot R \cdot d\phi \). The adhered bacteria also orientated with respect to local flow direction with a random angle of \( \theta \).
Figure 7.3 Free body diagram of a single adhered bacterial cell. Bacterial cell is simply assumed to be cylindrical. (a) The top view shows the flow direction “X”. $\theta$ is the inclination angle of bacteria against flow direction. Hydrodynamic drag force in X and Y direction were derived by COMSOL. Attachment and detachment moment along longitudinal direction and transverse direction were indicated. (b) Side view shows the free body diagram of bacteria. Pivot of moments is located at point “O”. Contact radius is $a$.

As bacteria adhering on a collector surface, the Figure 7.3 shows the free body diagram. The orientation angle $\theta$ in the Figure 7.3 (a) indicates the orientation of bacteria with respect to stream line on the collector surface. The drag force acting on a bacterium also depends on the orientation angle. For example, bacteria with larger orientation angle is expected to be detached by drag force more easily. The geometry defined above includes two critical parameters, $\phi$ and $\theta$. In summary, $\phi$ defines where the bacteria is located on a collector and $\theta$ defines the orientation of bacteria with respect to the flow. These two parameters determine the magnitude of hydrodynamic interaction applied on bacteria and thus affect the fate.
7.2.2.2 Forces

The external forces that bacteria attached on collector surface experience are shown in the Figure 7.3 (b), including the hydrodynamic drag, lift, gravity, buoyancy, the van der Waals, electrostatic double layer (EDL) and Hertz contact forces. The hydrodynamic drag force is introduced briefly in the previous section and will be discussed in detail in the “Flow Model” section. Our calculation indicated that effect of lift force is negligible compared to others. Measured density of wet bacteria is in a range of 110% ~120% of density of water (Carrera, Zandoneni, & Sagripanti, 2008). Consequently, bacteria tend to sink if no other external forces are exerted and the resultant of gravity and buoyancy forces is around 10~20% \( \rho g V \), where \( \rho \) is water density and \( V \) is bacterial volume.

The DLVO theory is applied to calculate the surface interaction between bacteria and collector surface as the superposition of van der Waals attraction (Gregory, 1981) and electrostatic double layer repulsion (Hogg et al., 1966) at ionic concentrations ranging from 1 to 10 mM. The intersurface interaction energy is determined by treating bacteria as an equivalent sized sphere and collector as a flat plate. The Pauli’s exclusion principle and the subsequent repulsive interaction are not considered in the present context. The DLVO potential is thus written as

\[
V(h) = -\frac{Aa_p}{6h(1+14h/\lambda)} + \pi \varepsilon_0 \varepsilon_r \alpha_p \left\{ 2\psi_\infty \psi_r \log \left( \frac{1+e^{-\psi_h}}{1-e^{-\psi_h}} \right) + (\psi_r^3 + \psi_\infty^3) \log[1-e^{-2\psi_h}] \right\}
\]  

(7-1)
Figure 7.4. DLVO interaction potential under ionic concentration of 1 mM, 3 mM and 10 mM. Repulsive barriers are from 80 to 700 kT and attractive well in the secondary minimum are from 0.1-2 kT. The figure in upper-left corner is zoom in for the secondary minimum. Primary minimum and Pauli repulsion are within 2 nanometers and not indicated in the figure.

where $A$ is the Hamaker constant of the interacting media (bacteria-water-quartz) (Redman et al., 2004), $a_b$ is the equivalent bacterial radius, $h$ is the separation distance between bacterial surface and collector surface, $\lambda$ is the characteristic wave length of the dielectric (Redman et al., 2004), $\epsilon_0$ is the dielectric permittivity in vacuum, $\epsilon_r$ is the relative dielectric permittivity of water, $\kappa$ is the inverse Debye length and $\psi_p$ and $\psi_c$ are the surface potential of the bacteria and quartz collector. The DLVO potential possesses two finite energy wells, primary minimum (1min) and the secondary minimum (2min), and a repulsive energy barrier ($R$), as shown in Figure 7.4. To simplify the involved mathematical manipulation, a stepwise Dugdale-Maugis approximation is adopted. The disjoining pressure then takes the form of a step-wise distribution,
\[
p(z) = \begin{cases} 
0 & \text{for } 0 < z \leq z_0 \text{ and } 0 < s < a \\
p_1 & \text{for } z_0 < z \leq z_1 \text{ and } a < s \leq c_1, \text{1 min} \\
p_R & \text{for } z_1 < z \leq z_R \text{ and } c_1 < s \leq c_R, \text{energy barrier} \\
p_2 & \text{for } z_R < z \leq z_2 \text{ and } c_R < s \leq c_2, \text{2 min} \\
0 & \text{for } z > z_2 \text{ and } s > c_2 
\end{cases} 
\]

as shown in Figure 7.5. Here \( z \) is the separation distance between the bacterial surface and the collector surface, \( z_0 \) is the equilibrium spacing between the two surfaces, \( z_1, z_R \) and \( z_2 \) are the heights of the three interaction layers from the sand surface, \( s \) is the radial distance of the bacterial profile from the polar axis, \( a \) is the contact radius and \( c_1, c_R, c_2 \) and \( s \) are the radii of the three interaction layers. For further convenience, the radial distance \( s \) is normalized with respect to the contact radius \( a \) as \( \xi = s/a \). The disjoining pressure then becomes

\[
p(z) = \begin{cases} 
0 & \text{for } 0 < z \leq z_0 \text{ and } 0 < \xi < 1 \\
p_1 & \text{for } z_0 < z \leq z_1 \text{ and } 1 < \xi \leq \xi_1, \text{1 min} \\
p_R & \text{for } z_1 < z \leq z_R \text{ and } \xi_1 < \xi \leq \xi_R, \text{energy barrier} \\
p_2 & \text{for } z_R < z \leq z_2 \text{ and } \xi_R < \xi \leq \xi_2, \text{2 min} \\
0 & \text{for } z > z_2 \text{ and } \xi > \xi_2 
\end{cases} 
\]

as shown in Figure 7.5.

In summary, all forces are classified into two categories, the detachment force that tend to detach the bacteria from the collector surface and the attachment force that tend to retain the bacteria. The detachment force only includes the hydrodynamic drag.
force. The total attachment force includes the resultant of the gravity and buoyancy forces, the integral of the DLVO pressure in the cohesive zone and the integral of the adhesive stress within the contact area. To derive the attachment force and contact radius, a cylindrical contact model is adopted.

7.2.2.3 Cylindrical Contact Model

A cylindrical contact model with one layer of constant adhesive stress based on Maugis analysis was derived by Johnson and Greenwood in 2008 (K. Johnson & J. Greenwood, 2008). In the present study, this model is extended by considering three layers of cohesive zones, as shown in the Figure 7.5. The force equilibrium in vertical direction (per unit length) is

$$ P = \frac{\pi E a^2}{4R} \left[ \text{Resultant of Gravity and Buoyancy} \right] + \frac{(\Delta \rho) g \pi R^2}{\text{Adhesion in contact}} + F_{s, \text{inside}} + F_{s, \text{outside}} \quad (7-4) $$

where $P$ is the applied load, and $F_{s, \text{outside}}$ is the adhesive load outside the contact region per unit length that is given as follows

$$ \frac{F_{s, \text{outside}}}{a} = -p_1 (\xi_1 - 1) + p_R (\xi_R - \xi_1) - p_2 (\xi_2 - \xi_R) \quad (7-5) $$

The adhesive stress distribution within the contact area can be assembled using the Green’s function (K. Johnson & J. Greenwood, 2008; Tada, Paris, & Irwin, 1985; Westergaard, 1939)

$$ g(\zeta, \xi) = \frac{a}{\pi} \sqrt{1 - \frac{\zeta^2}{\xi^2}} \left( \frac{1}{\xi - \xi_2} + \frac{1}{\xi + \xi} \right) \quad 0 < \zeta \leq 1, \ 1 < \xi < \xi_1 $$

$$ \sigma_1(\zeta) = \int_{\xi_1}^{\xi} p_1 g(\zeta, \xi) d\xi, \quad 0 < \xi \leq 1, \ 1 < \zeta < \zeta_1 \quad (7-7a,b,c) $$

$$ \sigma_R(\zeta) = \int_{\zeta}^{\xi_R} p_R g(\zeta, \xi) d\xi, \quad 0 < \zeta \leq 1, \ \zeta_1 < \zeta < \zeta_R $$

$$ \sigma_2(\zeta) = \int_{\xi_2}^{\zeta} p_2 g(\zeta, \xi) d\xi, \quad 0 < \zeta \leq 1, \ \zeta_R < \zeta < \zeta_2 $$

Note that the negative sign in Equation (7-7a,b,c) indicates the repulsion in the repulsive layer. The adhesive load within the contact (per unit length) is thus derived by
The displacement due to adhesive stress outside the contact area was ignored in the present study. The gap between the cylinder and substrate follows the Hertz contact theory as,

\[ H(\xi) = \frac{a^2}{2R} \left[ \frac{1}{2} (\xi^2 - 1) - \cosh^{-1} \xi \right] \]  

(7-9)

Thus, the condition for gap at \( \xi = \xi_1, \xi_R \) and \( \xi_2 \) to be the height of 1min layer \( z_1 \) is

\[
\begin{align*}
H(\xi) &= z_1, \quad 1 < \xi \leq \xi_1 \\
H(\xi) &= z_R, \quad \xi_1 < \xi \leq \xi_R \\
H(\xi) &= z_2, \quad \xi_R < \xi \leq \xi_2 
\end{align*}
\]  

(7-10)

When applied load, \( P=0 \), substitute Eq (7-5) and Eq (7-8) into Eq (7-4), the force equilibrium equation is written as

\[
0 = \frac{\pi a^3}{4} \left( \Delta \rho \right) g \rho R^2 - p_1 (\xi_1 - 1) + p_R (\xi_R - \xi_1) - p_2 (\xi_R - \xi_2) + \int_0^1 (\sigma_1 + \sigma_R + \sigma_2) d\xi 
\]  

(7-11)

Four unknowns, \( a, \xi_1, \xi_R \) and \( \xi_2 \) can be derived by solving Equations (7-9), (7-10) and (7-11). The total attachment force, \( F_a \), exerted on bacteria and contact radius of bacteria are thus derived.

7.2.2.4 Flow Model

As mentioned in section of “Geometry”, the drag force on the collector, based on Happel’s sphere-in-cell model (Happel, 1958; Shen et al., 2010), only depends on \( \phi \), shown in the Figure 7.2 as

\[
F_h(\phi) = F_h(\phi = 90^\circ) \times \sin(\phi) 
\]  

(7-12)

The maximum hydrodynamic drag force experienced by bacteria is at the center top of collector, \( \phi = 90^\circ \) and the minimum drag stress occurs at stagnation points, \( \phi=0^\circ \) and \( \phi=180^\circ \). \( F_h(\phi=90^\circ) \) is calculated by computational fluid dynamics. The commercially available finite element package COMSOL (Burlington, MA) was used in simulations. The geometry in COMSOL was set to as a rectangular channel, representing the void between the cubic packed sand grains. Dimension was chosen based on the experimental set up. The boundary conditions are: (i) inlet velocity is set to be interstitial velocity in the test (ii) outlet pressure is zero (iii) channel walls have no slip conditions. A cylinder with bacterial size is confined on the channel bottom with varying inclination angles \( \theta \). The main coordinate axis is “X” pointing in the flow
direction and the “Y” axis is perpendicular to the flow direction. The auxiliary coordinate on the bacterium with “x” axis points in the longitudinal direction of bacterium and the “y” axis points in the transverse direction, shown in Figure 7.3 (a). The angle between the two coordinate systems is $\theta$. The hydrodynamic drag force components on the bacterium in the “X” and “Y” directions are computed by COMSOL. Thus, the hydrodynamic drag forces may be converted to auxiliary coordinate system as follows,

$$
\begin{bmatrix}
F_{h,x} \\
F_{h,y}
\end{bmatrix} =
\begin{bmatrix}
\sin \theta & \cos \theta \\
-\cos \theta & -\sin \theta
\end{bmatrix}
\begin{bmatrix}
F_{h,x} \\
F_{h,y}
\end{bmatrix}
$$

(7-13)

Expressing the drag force in an auxiliary coordinate system is practical because the incipient rolling occurs locally at either longitudinal direction or transverse direction of the bacterium. Thus far, the hydrodynamic drag force, or the detachment force is derived.

7.2.2.5 Moment Balance Model

Three incipient motions of bacterial detachment are rolling, sliding and lifting. Although system dependent, rolling is often the predominant detachment mechanism for hydrodynamic detachment of colloids (Bergendahl & Grasso, 2000; Torkzaban et al., 2007). Incipient rolling occurs when detachment moment overcomes the attachment moment. The rolling pivot is assumed to be located at the front edge of contact, shown as point “O” in the Figure 7.3 (b). For single bacterium, rolling occurs at either longitudinal or transverse direction. This depends on along which direction the detachment moment first overcomes the attachment moment. For example, when bacteria are aligned with the stream, the hydrodynamic drag force in the “y” direction is zero, thus rolling along the longitudinal direction is expected to occur first. As the inclination angle $\theta$ increases, the hydrodynamic drag moment in the “y” direction increases significantly and rolling in transverse direction starts to dominate. By using the attachment and detachment forces derived in the previous sections, the attachment moment in longitudinal and transverse directions are found as follows,

$$
\begin{bmatrix}
M_{a,x} \\
M_{a,y}
\end{bmatrix} = F_a \cdot \frac{a}{L/2}
$$

(7-14)

The detachment moment, or the hydrodynamic moment in the auxiliary coordinate (local coordinate) were derived as shown in the Figure 7.3 (b),
\[
\begin{bmatrix}
M_{b-x} \\
M_{b-y}
\end{bmatrix} = \begin{bmatrix}
F_{b-x} \cdot a \\
F_{b-y} \cdot L/2
\end{bmatrix}
\]  
(7-15)

The direction of the moments follows the right-hand rule. For example, moment in “x” direction is calculated by force in “y” direction. With attachment moment and detachment moment, the detachment for single bacteria can be determined by using the following relationship,

\[
\max \left( \frac{M_{b-x}}{M_{a-x}}, \frac{M_{b-y}}{M_{a-y}} \right) \geq 1
\]  
(7-16)

### 7.2.2.6 Distribution of the Bacterial Concentration on the Collector Surface

The bacterial concentration distribution around a collector is determined by the following convection-diffusion equation \((1-2)\) (Elimelech, 1994),

\[
\nabla \cdot (uC) = \nabla \cdot (D \cdot \nabla C) - \nabla \left( \frac{D \cdot F}{kT} C \right)
\]  
(7-17)

where \(u\) is the flow velocity, \(C\) is the concentration of the bacteria, \(D\) is the particle diffusion tensor, \(k\) is the Boltzmann constant, \(T\) is the absolute temperature, and \(F\) is the external force vector. Using finite different method, Equation \((7-17)\) was solved and bacterial concentration “\(C\)” is derived as a function of \(\phi\) and separation \(H\), distance from bacteria to collector surface. The author followed the same scheme and mesh technique used in Elimelech’s work in 1994 (Elimelech, 1994). Detailed derivation of scheme and MATLAB code are given in the Appendix 3. Bacterial concentration distribution in vicinity of collector surface is thus derived by \(C(\phi, 0+\Delta H)\) where \(\Delta H\) is sufficiently small distance from collector surface. The bacterial density distribution on collector surface is then written as \(C(\phi)\).

At a given location \(\phi\), a circular latitudinal region is determined, where the hydrodynamic stress is constant, indicated by the grey area in the Figure 7.2. In this region a threshold value of the orientation angle, \(\theta^*\), can be found by solving Equation \((7-16)\). If \(\theta > \theta^*\), detachment occurs and if \(\theta < \theta^*\) attachment occurs. \(\theta^*\) is a function of \(\phi\), expressed as \(\theta^*(\phi)\). Distribution of the orientation angle of the bacteria on the collector surface is measured by the microfluidic test. The ellipsoidal particles are prepared by stretching of spherical polystyrene particle (Yao et al., 1971). The parallel plate flow chamber was first filled with 60 mM Potassium Chloride (KCl) solution as the background solution. Then 60 mM KCl solution with suspended ellipsoidal polystyrene particles was injected at a low average flow rate of 0.015 mm/s, using a syringe pump.
A converted microscope was adopted to focus on the particles in the vicinity of the chamber substrate. After the system reached the steady state, snapshot of the particles was taken. Thus, the orientation angle distribution on the collector surface was obtained after image processing for elongated polystyrene particles. The distribution of the orientation angle at a given $\phi$ is expressed as $G(\theta,u)$ where $u$ is flow velocity.

### 7.2.2.7 Model for attachment efficiency $\alpha$

Since the distribution of the bacterial density on the collector surface, $C(\phi)$ and the threshold inclination angle for local region $G(\theta,u)$ are found, the attachment efficiency for a single collector $\alpha$ can be obtained by,

$$\alpha = \frac{\int_{0}^{\pi} G(\theta,u) \cdot C(\phi) 2\pi R \sin \phi \cdot Rd\theta \cdot d\phi}{\int_{0}^{\pi} C(\phi) 2\pi R \sin \phi \cdot R \cdot d\phi} \quad (7-18)$$

Using Eq (7-18), the attachment efficiency in the primary minimum $\alpha_1$ and secondary minimum $\alpha_2$ are calculated respectively. In this work, the fraction of bacteria “jump” into primary minimum $f_1$ and retain in the secondary minimum $f_2$ is derived by finding the best fit between column test results and modeling results. The overall attachment efficiency can be calculated as,

$$\alpha = \alpha_1 f_1 + \alpha_2 f_2 \quad (7-19)$$

As a summary, a scheme of the new model and the CFT theory is shown in the Figure 7.6. It shows the overall mechanism of the new method and difference with the CFT theory. The parameters considered in the both methods are also listed.
Figure 7.6. Scheme of the new method and CFT theory. Blue arrows and boxes are the new model. The red arrows and boxes are the CFT theory. The black arrows and boxes it connects are shared by the new model and CFT theory. From the scheme note that the main difference between the new model and CFT theory is that: new model uses the moment compare method to calculate rate of bacteria attach on a sand and CFT theory uses the experiment. Both methods use the adopt the similar theory to calculate the rate of bacteria collide with a sand.

7.3 Results and Discussion

7.3.1 Adhesive moment and hydrodynamic moment

Adhesive moments in primary and secondary minimum for each strain are calculated for all tested ionic concentrations in the longitudinal and transverse directions. Note here that the direction for moment and rolling follows the righthand rule. Results are shown in the Table 7.1. Only hydrodynamic moment at location $\varphi = 90^\circ$, center top of the collector, is listed at varying flow rates. Because the hydrodynamic moment at other locations can be calculated as, $M_{hy}(\varphi) = M_{hy}(90^\circ)\sin\varphi$, according to sphere in cell theory. Note that the range of hydrodynamic moment reflects varying flow rates and bacterial orientation.
### Table 7.1 Adhesive moment (mN)

<table>
<thead>
<tr>
<th>Bacterial type</th>
<th>Ionic concentration</th>
<th>Adhesive moment in primary min</th>
<th>Adhesive moment in secondary min</th>
<th>Hydrodynamic moment</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Longitudinal</td>
<td>Transverse</td>
<td>Longitudinal</td>
</tr>
<tr>
<td>Strain A</td>
<td>1mM</td>
<td>3.61×10^{-19}</td>
<td>3.43×10^{-17}</td>
<td>8.69×10^{-22}</td>
</tr>
<tr>
<td></td>
<td>3mM</td>
<td>3.67×10^{-19}</td>
<td>3.49×10^{-17}</td>
<td>5.46×10^{-22}</td>
</tr>
<tr>
<td></td>
<td>10mM</td>
<td>5.76×10^{-19}</td>
<td>4.50×10^{-17}</td>
<td>2.63×10^{-20}</td>
</tr>
<tr>
<td>Strain Q</td>
<td>1mM</td>
<td>5.81×10^{-19}</td>
<td>3.85×10^{-17}</td>
<td>7.37×10^{-22}</td>
</tr>
<tr>
<td></td>
<td>3mM</td>
<td>6.88×10^{-19}</td>
<td>3.56×10^{-17}</td>
<td>4.25×10^{-21}</td>
</tr>
<tr>
<td></td>
<td>10mM</td>
<td>1.06×10^{-18}</td>
<td>4.13×10^{-17}</td>
<td>2.33×10^{-20}</td>
</tr>
</tbody>
</table>

Table 7.1 Adhesive moments and hydrodynamic moment derived by calculation under varying ionic concentrations and flow rates. Range of hydrodynamic moment is corresponding to the range of flow rate from 5 mL/min to 50 mL/min.
The adhesive moment in the secondary minimum is two orders of magnitude smaller than the hydrodynamic moment. This indicates that bacteria rarely attach in the secondary minimum. For attachment in the secondary minimum to occur the following conditions should be satisfied: (i) bacteria located in the vicinity of the stagnation point, (ii) low flow rate, (iii) high ionic concentration, (iv) bacteria aligned with the streamline. All these conditions are reflected by the lower end of the hydrodynamic moment in the Table 7.1. The magnitude of the adhesive moment in the primary minimum is comparable to the hydrodynamic moment. Thus, attachment of bacteria more likely occurs in the primary minimum and is subjected to flow rate. Because adhesive moment in the transverse direction exceeds the hydrodynamic moment by over one order of magnitude due to long lever arm, detachment in the primary minimum is more likely to happen in the longitudinal direction. Discussion of the threshold orientation angle in the next section is based on the calculation in the primary minimum. The adhesive moments derived in this study are around 3 orders of magnitude higher than adhesive moments derived by Shen (Shen et al., 2010). This is because Shen used spherical particle in his study thus the contact area and adhesive force were different.

### 7.3.2 Threshold orientation angle

Given the ionic concentration and the bacterial type, the adhesive moment of a single bacterium is determined. Given the flow rate and the location $\phi$, the hydrodynamic moment acting on a single bacterium is also determined. By comparing the adhesive moment to the hydrodynamic moment, the threshold inclination angle is obtained by Equation (7-7a,b,c) as a function of the flow rate, location $\phi$ on the collector with varying ionic concentration. Results are shown in the Figure 7.7 for three different bacterial strains. Different ionic concentrations are indicated by different colors as the flow rate varies from 5 mL/min to 50 mL/min. According to the sphere-in-cell model, the hydrodynamic stress is symmetrically distributed around $\phi = 90^\circ$. Thus, only $\phi = 0^\circ$~$90^\circ$ are calculated here. Location positions $\phi = 0^\circ$ and $\phi = 90^\circ$ indicate the front stagnation point and center top of the collector respectively, as shown in the Figure 7.2. Threshold angle $\theta = 90^\circ$ indicates at given flow rate and location, circular latitudinal region shown in Figure 7.2, all bacterium attached successfully. Threshold angle $\theta = 0^\circ$ indicates at given flow rate and location all bacterium is removed by fluid drag interaction.

### 7.3.3 Effects of Flow Rate
Take strain A for instance, indicated by Figure 7.7 (a), at the low flow rate of 5 mL/min, all bacteria are retained on the collector indicated by the threshold angle of $\theta = 90^\circ$ at all locations. As flow rate increases, detachment starts to occur at the area close to collector center top. When the flow rate reaches maximum value of 50 mL/min, all bacteria in the vicinity of the front stagnation point, or location angle $\phi$ is smaller than 10’, are retained. Thus, for each flow rate, a critical location angle exists when the location angle is smaller than the critical location angle, bacterium always remain stuck. Otherwise, attachment depends on the threshold angle. The critical angle is clearly a function of flow rate as shown in the Figure 7.7. Critical location angles decrease from 90’ to 10’ for strain A as the flow rate increases from 5 mL/min to 50 mL/min. Strain Q and strain H are more adhesive compared to strain A, indicated by the higher threshold angle. For example, critical location angle is 90’ for strain Q at 5,10,15 mL/min. No detachment occurs for strain Q under 15 mL/min. This explains how flow rate affects the bacterial attachment efficiency. High flow rate lowers the attachment efficiency by decreasing the overall threshold angle.
Figure 7.7. Threshold inclination angle, $\theta$ for strain A, Q and H under varying ionic concentration and flow rate on a collector. Higher inclination angle indicates higher attachment efficiency. $\theta = 90^\circ$ suggests that at the circular latitudinal region determined by corresponding location $\phi$, all bacteria are attached.

### 7.3.4 Effects of Ionic Concentration

According to DLVO theory, higher ionic concentration decreases repulsive energy barrier caused by electro-static repulsion and results in deeper primary and secondary minimum potential, as shown in the Figure 7.4. In Figure 7.7, this is reflected by the higher threshold angle at local $\phi$ for higher ionic concentration. Recall that Figure 7.7 only shows the results for primary minimum potential because secondary minimum potential is so shallow that the threshold angle for all location and flow rate is expected to be zero. Results show that the threshold angle for 1mM and 3mM KCl solution are
similar, indicated by red and blue curve in the Figure 7.7. The threshold angle for 10 mM KCl solution is higher than the other two ionic concentrations for all three strains indicated by the black curves in the figure.

Thus, the effects of variation of the ionic concentration on the threshold angle is not significant. This is because in the primary minimum layer, the van der Waals force dominates. The electro-static repulsion is a long range force. It vanishes in the primary minimum. Note that column test shows an apparent effect of the ionic concentration on the filtration efficiency. Thus, we conclude that ionic concentration determines the ratio of bacteria that go into the primary minimum. Once bacteria fall into the primary minimum, the effect of ionic concentration is very limited. Fraction of bacteria that go into primary minimum can be obtained by fitting calculation results with column test. This will be discussed in detail in the next section.

7.3.5 Attachment Efficiency

By definition, attachment efficiency is the ratio between number of bacteria that attach on the collector to the number of bacteria that hit the collector. This is expressed by Equation (7-8). To solve Eq (7-8), three key parameters are needed: threshold inclination angle $\theta_{\text{threshold}}$, inclination angle distribution $G(\theta, v)$ and the bacterial concentration distribution $C(\phi)$ in vicinity of collector surface. Threshold inclination angle is a function of location angle $\phi$ and flow rate $v$ as discussed in the previous section. Bacterial concentration distribution $C(\phi, H)$ was derived by many former researchers, e.g. Elimelech, by solving the classic convection-diffusion equation using finite difference scheme (Elimelech, 1994). In this work, the same method was adopted. The bacterial density distribution derived in this study is shown in the Figure 7.8. The bacterial concentration distribution around the collector surface is selected from the distance of 100 nanometer away from the collector surface as $C(\phi, H=100 \text{ nm})$. This distance is picked because the DLVO force range is around 100nm according to our calculation. Once bacterium fall into this region surface interaction start to affect and the moment comparison mechanism is initiated.
Figure 7.8 Dimensionless bacterial concentration distribution around a spherical collector. $\phi$ is tangential coordinate and $H$ is perpendicular coordinate, start from collector surface. This result was derived under 3mM of KCL and flow rate of 10 mL/min.
Microfluidic test is applied to measure the orientation angle distribution of elongated polystyrene particles $G(\theta,v)$. Results suggest a Gaussian distribution and show a likely correlation between the flow rate and the distribution parameters, as shown in the Figure 7.9. The tests were conducted only on flow rate of 5 mL/min and 50 mL/min as shown in the Figure 7.9. The standard deviation, $\sigma$, corresponding to these two flow rates are 40° and 75°. For the flow rate in range of 5-50 mL/min, we assume a linear relationship between $\sigma$ and flow rate. Higher flow rate corresponds to narrow standard deviation. In other words, around 70% of elongated particles on collector form an angle smaller than 40° to 75° with respective to streamline. Although the correlation between flow rate and distribution is implicit due to lack of enough experimental data, a further pursue of accurate correlation equation of flow rate and distribution parameter is out of scope of this dissertation. Thus, the attachment efficiency in the primary minimum, $\alpha_1$ and the secondary minimum, $\alpha_2$ can be obtained by Eq (7-8), respectively.
Figure 7.9. Measured inclination angle under flow rate of 5 and 25 mL/min. Number of bins are set at 60 for range from -90° to 90°.

The fraction of bacteria that reaches the primary minimum $f_1$ and secondary minimum $f_2$ are determined by fitting the theoretical results and column test results. Ratio of bacteria that are able to overcome the energy barrier and reach the primary
minimum increase as ionic concentration increases. Results are shown in the Table 7.2. Under ionic concentration of 1mM and 3mM, the ratio of the bacteria that reach the primary minimum are 29% and 40% respectively for both strains. Under ionic concentration of 10 mM, 53% of strain Q are able to reach the primary minimum and achieve strong attachment. 43% of strain A are able to reach the primary and achieve strong attachment. Due to the high energy barrier calculated by the classical DLVO theory, almost zero fraction of particle reaching the primary minimum are always predicted under low ionic concentration. In 2004, Tufenkji experimentally showed that a fraction of polystyrene particle population, 5%, deposited in the primary minimum under low ionic concentration, 3 mM of KCl. She explained this is due to the surface charge heterogeneity of the particle and the collector (Tufenkji & Elimelech, 2004b). In this study, the surface charge heterogeneity also occurred on the bacterial and collector surface therefore a fraction of bacteria experience small energy barrier. Our results indicate even higher primary deposition ratio than Tufenkji predicted. We believe that this is mainly due to the biological complexity of bacteria such as the bacterial motility and bacterial surface substances. For example, one of explanations is the surface appendages of bacteria are folded under high ionic concentration (X. Wang et al., 2012). This potentially enclose the distance between bacteria membrane and collector surface.

<table>
<thead>
<tr>
<th>Species</th>
<th>1mM</th>
<th>3mM</th>
<th>10mM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Strain A</td>
<td>0.29</td>
<td>0.41</td>
<td>0.43</td>
</tr>
<tr>
<td>Strain Q</td>
<td>0.30</td>
<td>0.41</td>
<td>0.53</td>
</tr>
</tbody>
</table>

Table 7.2 Fraction of bacteria, strain A and strain Q, reach the primary minimum at three ionic concentrations.

Overall attachment efficiency derived by Equation(7-10) is shown in the Figure 7.10. In spite of high fraction of bacteria reaching secondary minimum, from 50% to 70%, successful attachment in this region is very rare due to weak adhesion. Results in the Figure 7.10 are largely due to the attachment in the primary minimum. Scattered points are results derived by combining column test results and colloidal filtration theory and solid curves are results derived from the model in this study. Two results agree very well. Therefore, the theoretical model in this study captured the information
provided by the column test in the conventional method. In other words, the
determination of particle attachment and detachment after it collides the collector are
now provided by our physical model, comparing the removal moment and adhesive
to moment, instead of column test.

![Graph showing filtration efficiency for strains A and Q](image)

Figure 7.10. Filtration efficiency derived by the new model and column test for strain A and Q.

### 7.4 Conclusion

Conventional CFT method of predicting attachment efficiency is a combination
of theoretical modeling and experimental method. Some crucial factors, for example,
bacterial/colloidal geometry, hydrodynamic interaction, electrostatic interaction and
elastic modulus, are mined in the experimental results. The attachment/detachment
 mechanism has not been clear. The model presented in this study assumes the
attachment/detachment mechanism by using the moment balance approach. It includes the effect of bacterial elastic modulus, geometry, hydrodynamic interaction and bacteria-collector surface interaction. The calculation results agree well with the column test results. Attachment efficiency of strain Q is higher than that of strain A. We believe this is because the contact area of strain Q with the collector is larger compared to strain A due to softness of strain Q. Effect of flow rate is captured. Coupled effect of flow rate and ionic concentration can be explained. Higher flow rate causes higher drag moment and thus lower attachment efficiency. For the bacteria in the secondary minimum, adhesion for three ionic concentrations are all very weak compared to hydrodynamic interaction tested in this study. Attachment in the secondary minimum is negligible. Adhesion in the primary minimum is dominated by van der Waals interaction and the magnitude of the adhesive interaction is comparable to the hydrodynamic interaction. Thus, the flow rate influences bacterial detachment in the primary minimum.

This model does not include a theoretical method to determine fraction of bacteria jump into the primary minimum and secondary minimum. We empirically derived this fraction by fitting the calculation results and experimental results. The fraction predicted in this study, 30% to 50%, are higher than fraction derived by using the thermal fluctuation method (Rijnaarts et al., 1996b), kinetic energy method (M. W. Becker et al., 2004; Rijnaarts et al., 1995) and dual deposition model (Tufenkji & Elimelech, 2004b). We believe this is due to biological complexity of bacteria.

Overall, the moment compare model explains the attachment/detachment mechanism after collision very well. It shows a great potential to be a platform for further study on more complex system including more factors missing in this study, for example, bacterial motility and effects of microbial surface substances.
Chapter 8. Conclusions and Future Work

8.1 Conclusions

The preliminary work showed a strong correlation between micro-mechanical properties of bacterium and their macro-filtration behavior (Y. Li et al., 2014). This motivated us to incorporate the fundamental surface science and solid mechanics into the subject of microbial adhesion-transportation, potentially improving the conventional empirically driven approach for predicting microbial attachment and transportation in porous medium.

This dissertation focuses on microbial and colloidal transportation and filtration in porous medium in aqueous environment using a new microfluidic method. The new method reduced the complexity of the problem and allowed us to differentiate individual factors involved. The colloidal attachment and detachment were firstly studied with a wide range of flow rates and ionic strengths. Coupled effect of flow rate and ionic strengths were analyzed based on the classic DLVO theory. The fundamental surface science was revealed and a theoretical model was built based on new findings. Then four types of bacterium associated with human health and water pollution were investigated using the new microfluidic method. Lastly, a new theoretical model for bacterial filtration in porous medium were established based on a moment balance method, namely the fate of bacterium retained on a collector is determined by the competition between detachment moment due to hydrodynamic interaction and attachment moment due to surface adhesion.

In chapter 3, a simple adhesion model is derived for an elastic sphere adhering to a rigid planar substrate in the presence of moisture. The model is useful in discussing adhesion of microscopic objects, and the derived values of “pull-off” force and contact radius can be used to deduce the magnitude and range of inter-surface forces. Contrary to the classical description based on Maugis’s JKR-DMT transition, meniscus alone causes $|F^†|$ to exceed $2\pi R\gamma$ for any relative humidity and the DMT limit only holds at saturation. It also emphasized that once RH drops to roughly 30%, water condensation and consequent adhesion in glass become relative unimportant. Another major outcome of this work is to decouple the capillary force from the solid-solid interaction.

Although our preliminary work already showed that micro-mechanical properties of bacterium is correlated to their filtration behavior, the impact of fluid velocity and coupled effect of flow rate, bacterial species and ionic strength are yet to
In chapter 4, handful column tests were conducted on 3 types of bacterium with a wide range of flow rate (5 to 50 mL/min) and a wide range of ionic strengths (1, 3 and 10 mM KCl). Filtration efficiency of strain A and strain Q were negatively correlated to flow rate and were positively correlated to ionic strength. Based on DLVO theory, we conducted a qualitative analysis on surface interaction between bacterium and collector surface under varying ionic strength. Higher ionic strength cause more bacterium fall into the 1st min. The impact of flow rate can be explained by the competition between hydrodynamic interaction and DLVO attraction. The magnitude of attraction in the 1st min is comparable to hydrodynamic interaction. The magnitude to attraction in the 2nd min is negligible comparing to hydrodynamic interaction. Thus, under 1 mM of KCl, almost all bacterium retained were in the 2nd min and the attachment was so weak that detachment was not subject to the flow rate. While under 10 mM of KCl, the detachment was subject to the flow rate. The modified Tabor’s parameter of \( \mu \) was found to be a function of flow rate.

In chapter 5, a new microfluidic method was invented to investigate adhesion-detachment of colloidal polystyrene particles in an electrolyte in the presence of flow. A theoretical model was constructed based on the classical DLVO theory which comprises a double energy well. While the 1st min, or primary minimum adhesion prevents particles from escaping, the weak secondary minimum determines the dependency upon coupled flowrate and ionic concentration. When the solution salt concentration exceeds a critical threshold of 60 mM, flowrate no longer has an effect on the particle adhesion-detachment, because majority of the particles are trapped in the primary minimum. The new theoretical model sheds lights on the underlying physics of filtration. The results have significant impacts on in-situ or enhanced subsurface bioremediation, drinking water supplies, and water / wastewater treatments. The microfluidic device provides an economical and simple way to investigate live bacterial cells.

In chapter 6, the new microfluidic test was applied to bacterium. Four bacterial strains associate with human health and water treatment were studied. Firstly, a good agreement between results of microfluidic test and results of column test was observed, shown in the Figure 6.5. We believe the new microfluidic test method has a great potential to take place of column test when experiment meets the following two conditions: (i) measuring filtration efficiency when particle size is much smaller than void size (ratio of diameter < 1:50) (ii) surface absorption is the main filtration
mechanism. Microfluidic test provides a direct way to measure filtration efficiency and is more time efficient compare to conventional empirical driven approach. The microfluidic test also captured the couple effect of bacterial properties (Tabor’s parameter) and flow condition on filtration efficiency. A dynamic Tabor’s parameter, a combination of bacterial properties and flow condition was developed. Figure 6.8 showed a strong correlation between bacterial filtration efficiency and the dynamic Tabor’s parameter. A special case was strain H. Filtration efficiency of strain H is not sensitive to flow velocity because its’ surface adhesion is so strong that it dominated bacterial detachment under all flow velocities. Meanwhile, strain A, Q and SH2 exhibited a low surface adhesion and high elastic modulus. Their detachment ratio was subject to their micro-properties and flow rate. Lastly, we investigated effect of inclination angle of bacterial settlement on their later-on detachment under flow. We found that detachment of strain Q and strain A randomly occurred in a span of inclination angle from 0° to 90°. For strain H, bacterial detachment rarely occurred due to strong surface adhesion. However, bacterium of strain H would orientate themselves and align with flow direction in the presence of hydrodynamic interaction. This further enhanced attachment of strain H on collector in filtration process.

In chapter 7, using moment balance method, we established a theoretical model on bacterial filtration in porous medium. The new model explains the attachment/detachment mechanism after collision very well. It shows a great potential for further study on more complex system including more factors missing in this study, for example, bacterial motility and effects of microbial surface substances.

8.2 Future work

This dissertation showed the promising microfluidic test method on microbial/colloidal transportation-adhesion behavior and filtration from porous medium. A number of promising future research directions are proposed based on this dissertation.

Firstly, we already conducted microfluidic tests on spherical polystyrene. In next step, we could look into irregular shaped particles, e.g. ellipsoidal particles. The correlation between filtration efficiency and aspect ratio of particles would be of interested to many disciplines, e.g. drug delivery, water treatment and food processing. We could also look into particle of modified surface chemistry. Thus, we are able to study the interaction beyond van der Waals force and EDL repulsion in aqueous
environment. We already conducted a few microfluidic tests on PVA coated polystyrene particles. The adhesion energy of particle surface was found higher than uncoated ones.

Secondly, although the attachment and detachment of bacterium and particle in the presence of flow were well explained using the moment balance model, the impact of biological factors on bacterial filtration were not well understood yet. For example, pili and flagella exist on most bacterial cell wall and they usually enhance bacterial attachment and mobility. These would modify the adhesion in both 1st and 2nd min, and orientation angle of bacterial settlement. A modified adhesion model integrating the effect of surfactant could be attempted in the future.

Thirdly, microfluidic test generates a great amount of image and video, describing dynamic information of bacterial detachment, attachment and transportation in the presence of flow. We believe more valuable information are embedded in these data and yet to be dug out. For example, the history of individual bacterial detachment and attachment. Attachment pattern of each bacterium are different, for instance, some bacterial cells partially attached and some other bacterial cells fully attached on the channel surface. In the future, the image process program could be improved and applied to investigation of these aspects. We believe such a study would provide a better understanding of coupled influence of biological factors, flow condition and environments.

Lastly, microfluidic channels of complex structure could be designed fabricated in the future and are applied to mimic 2D version of 3D aqueous environment, for instance, underground porous medium and intestine/blood vessels in human body.
REFERENCES


APPENDIX 1. DERIVATION OF THE DLVO INTERACTION BETWEEN A SPHERE AND A FLAT SURFACE

The first of four field equations, called Maxwell’s equations, describes how electric fields emanate from electric charges and is written as,

$$\nabla \cdot E = \frac{\rho}{\varepsilon}$$  
(A1-8-1)

Here $\rho$ is electric charge density and $\varepsilon$ is dielectric constant of the medium. Electric field generated by a set of stationary charges can be written as the gradient of electric potential, so that

$$E = -\nabla \psi$$  
(A1-8-2)

Here $\psi$ is electric potential. Combining equation (A1-8-1) and (A1-8-2), give a very famous type of partial differential equation known as Poisson’s equation,

$$\nabla^2 \psi = -\frac{\rho}{\varepsilon}$$  
(A1-8-3)

Note that both $\psi$ and $\rho$ are a function of the spatial coordinate. Gouy-Chapman model extended application of equation (A1-8-3) to ionic solution but on basis of a number of further simplifying assumptions: ions are point charges of negligible dimension, surface charge is homogeneous spread and solvent is dielectric continuum. This is a first approximative theory for the electro double layer.

The ions density at a given point can be calculated from the electric potential at the same point with the aid of Boltzmann’s theorem:

$$n_- = n_0 \exp(\nu_- e \psi / kT)$$
$$n_+ = n_0 \exp(-\nu_+ e \psi / kT)$$  
(A1-8-4 a,b)

Here $n_0$ is bulk density of ions, $\psi$ is local electric potential, $e$ is the elementary charge, $\nu_-$ and $\nu_+$ are the valences of the positive and negative ions. For Potassium Chloride solution, the ions only own one valence and we therefore write $\nu_- = \nu_+ = \nu$. Electric Charge density $\rho$ is thus derived from equation (A1-8-4 a,b) as,

$$\rho = \nu e (n_+ - n_-) = -2n_0 \nu e \sinh(\nu e \psi / kT)$$  
(A1-8-5)

If the electric potential is so small that $\nu e \psi / kT << 1$ and $\exp(\nu e \psi / kT) \approx 1$, this may be approximated by $1 + \nu e \psi / kT$. This is known as Debye-Hückel approximation. We now
have an expression for the charge density, equation (A1-8-5), which may be inserted into equation (A1-8-3) and we thus obtain the fundamental differential equation:

\[ \nabla^2 \psi = \frac{2n_0 (ue)^2 \psi}{\epsilon kT} = \kappa^2 \psi \]  
(A1-8-6)

where \( \kappa \) is the inverse Debye-Hückel length and is given by

\[ \kappa^2 = \frac{2n_0 (ue)^2}{\epsilon kT} \]  
(A1-8-7)

For infinite flat surface, equation (A1-8-6) can be expressed in Cartesian co-ordinates as

\[ \frac{d^2 \psi}{dx^2} = \kappa^2 \psi \]  
(A1-8-8)

Solution of equation (A1-8-8) yields

\[ \psi = A_1 \cosh \kappa x + A_2 \sinh \kappa x \]  
(A1-8-9)

where \( A_1 \) and \( A_2 \) are constant to be determined.

For two dissimilar flat surfaces with distance of \( 2d \) apart, equation (A1-8-9) must satisfy the boundary conditions (i) \( \psi = \psi_{01} \) at \( x=0 \), and (ii) \( \psi = \psi_{02} \) at \( x=2d \), where \( \psi_{01} \) and \( \psi_{02} \) are surface potential of two surfaces. Applying the boundary conditions and equation (A1-8-9) may be written as

\[ \psi = \psi_{01} \cosh \kappa x + \left( \frac{\psi_{02} - \psi_{01} \cosh 2\kappa d}{\sinh 2\kappa d} \right) \sinh \kappa x \]  
(A1-8-10)

Equation (A1-8-10) indicates the distribution of electric potential as a function of distance between two flat surfaces.

**Potential energy of interaction between flat surfaces due to electric double layer**

To derive a quantitative expression for the potential energy \( V \) of interaction between flat surfaces, we need to obtain the free energy of each flat surface due to formation of electrical double layers. \( V \) is equal to the change in total free energy change of the double layer system when the two surfaces are brought together from infinity. As the free energy is identical with the amount of work to be performed in building up the double layers, the variation of free energy with surface distance directly equals the variation of the potential energy of two flat surfaces with respect to each other. Hence
the free energy can be derived by calculating work done by transferring ions from bulk solution to the surface.

The total work performed in building up the double layer system consists of two parts: chemical potential and electric potential. As s standard or reference state, we now take a neural surface with no adsorbed ions in contact with infinite bulk electrolyte, or ionic solution, containing potential determining ions of known fixed charge, \( q \), and initial chemical potentials, \( \mu_I \). For a single potential ion being adsorbed onto the surface, the free energy change can be written as

\[
g = (\mu_S - \mu_I) + q \psi'_0
\]

(A1-8-11)

Here \( \mu_S \) is chemical potential of an ion on the flat surface and \( \psi'_0 \) is the electrostatic potential on the surface at an arbitrary stage of double layer formation. We now write the number density of ions on the surface at an arbitrary stage of double layer formation as \( \Gamma \). The total free energy change per unit area of the surface may be written as

\[
G = \int_{\Gamma_0}^{\Gamma} \sigma \cdot d\Gamma = \int_{\Gamma_0}^{\Gamma} (\mu_S - \mu_I)d\Gamma + \int_{\Gamma_0}^{\psi'_0} q \psi'_0d\Gamma
\]

(A1-8-12)

Here \( \Gamma_0 \) is the number density of ions on the surface when final equilibrium has been established. If we write charge density on the surface at an arbitrary state and at final equilibrium state as \( \sigma \) and \( \sigma_0 \), the equation (A1-8-12) may be rewritten as

\[
G = \int_{\Gamma_0}^{\Gamma_0} (\mu_S - \mu_I)d\Gamma + \int_{\sigma_0}^{\sigma_0} \psi'_0(\sigma)d\sigma
\]

(A1-8-13)

Note the surface electric potential \( \psi'_0 \) is a function of \( \sigma \). We should bear in mind that Verwey and Overbeek made a further assumption here that chemical potential of an ion on surface \( \mu_S \) is a constant. Under high density of ions on the surface, \( \mu_S \) could depend on both the number of ions adsorbed and surface electric potential. Since only low electric potential was considered as mentioned in previous section, this assumption is still valid.

The first term in equation (A1-8-13) is called chemical part of the free energy. The second term is therefore identified as the electrical work done in creating the double layer. In evaluating the first integral in equation (A1-8-13), we recall that \( \mu_S - \mu_I \) is the free energy difference gained by a single ion due to chemical potential difference between the flat surface and bulk solution. In the final state of a single ion this chemical
free energy difference should exactly outweighs the electric potential difference and may be written as,

\[ q \psi_0^\delta (\sigma) + (\mu_s - \mu_i) = 0 \]  

(A1-8-14)

Here \( \psi_0^\delta(\sigma) \) is surface electric potential difference due to the work done by chemical potential. As assumed, setting \( \mu_s-\mu_i = \) constant is equivalent to setting \( \psi_0^\delta(\sigma) = \psi_0 \), a constant (see equation (A1-8-14)) so that from equation (A1-8-13) the free energy of the double layer becomes

\[ G = -\psi_0 \sigma_0 + \int_{\sigma_0}^{\sigma_f} \psi_0'(\sigma) d\sigma \]  

(A1-8-15)

To give a better physical sense of equation (A1-8-15), we can treat the double layer system simply as a spring-mass system. One end of the spring is fixed and the other end of the spring is subjected to mass with gravity of M. The initial state of the system is when the mass is connected to the spring but the gravity is not applied to the spring and spring is not elongated yet. Hence system is not at equilibrium at the initial state due to the gravity obviously. When mass is released, the gravity would drive the mass from the initial state to the final equilibrium state when the gravity is balanced out by the elongation of spring and spring is extended. If we treat the mass as a single ion, in this process, gravity plays a role as chemical preference and spring plays a role as electric potential. The first term in equation (A1-8-15) represent the work of the system under the gravity of mass on the spring, where \( \psi_0 \) represents the gravity and \( \sigma_0 \) represents the final extension of the spring. The second term in equation (A1-8-15) represents the work done by the spring, where \( \psi' \) is the spring force under an arbitrary extension, \( \sigma \).

When system reached final equilibrium, the electric surface potential is \( \psi_0 \) and surface charge density is \( \sigma_0 \). In formation of double layer, the free energy added to the system due to work done by chemical potential. The free energy added or subtracted from the system due to work done by electric potential. By partial integration both terms can be summarized into on single term

\[ G = -\int_{\sigma_0}^{\psi_0} \sigma(\psi_0') d\psi_0' \]  

(A1-8-16)
If the electric surface potential is small as assumed, the charge $\sigma$ and the electric potential $\psi_0$ increase proportionally, so that the linear approximation can be applied, equation (A1-8-16) simplifies to

$$G = -\frac{1}{2} \sigma_0 \psi_0$$  \hspace{1cm} (A1-8-17)

This linear relationship may be viewed as analogous to linear relationship between spring force and extension under small deformation in the spring system. Thus, the total free energy of the double layer systems of two dissimilar surfaces, with separation of $2d$, is equal to the sum of the free energies of the two separate surfaces and written as,

$$G_{12} (2d) = -(\frac{1}{2} \sigma_{01} \psi_{01} + \frac{1}{2} \sigma_{02} \psi_{02})$$  \hspace{1cm} (A1-8-18)

Here $\sigma_{01}$ and $\sigma_{02}$ are charge density on the two surfaces and $\psi_{01}$ and $\psi_{02}$ are the electric potential on the two surfaces at the equilibrium state. Recall that the potential energy of interaction between two surfaces is the change of total free energy in bringing two surfaces together from infinity. Hence the total potential energy density may be written as

$$V = G_{12} (2d) - G_{12} (\infty)$$  \hspace{1cm} (A1-8-19)

Recall that for infinite flat surface in ionic solution we have

$$\nabla^2 \psi = \frac{d^2 \psi}{dx^2}$$  \hspace{1cm} (A1-8-20)

Here $x$ is distance from the surface. Substitute equation (A1-8-20) into equation (A1-8-3) we derived $q$ as

$$\rho = -\varepsilon \left( \frac{d^2 \psi}{dx^2} \right)$$  \hspace{1cm} (A1-8-21)

Thus, by definition, the surface charge density may be derived by

$$\sigma_{01} = -\int_0^d \rho \cdot dx = \int_0^d \varepsilon \left( \frac{d^2 \psi}{dx^2} \right) \cdot dx = -\varepsilon \left( \frac{d \psi}{dx} \right)_{x=0}$$

$$\sigma_{02} = -\int_0^{2d} \rho \cdot dx = \int_0^{2d} \varepsilon \left( \frac{d^2 \psi}{dx^2} \right) \cdot dx = \varepsilon \left( \frac{d \psi}{dx} \right)_{x=2d}$$  \hspace{1cm} (A1-8-22 a, b)
Here \( \rho \) is spatial charge density and \( d \) is half distance between two surfaces where \( d \psi /dx = 0 \). Substitute equation (A1-8-10) into equation (A1-8-22 a, b) the surface charge density becomes

\[
\sigma_{01} = -\varepsilon \kappa \left[ \psi_{01} \tanh(2\kappa d) - \psi_{01} \coth(2\kappa d) \right] \\
\sigma_{02} = +\varepsilon \kappa \left[ \psi_{02} \tanh(2\kappa d) - \psi_{01} \coth(2\kappa d) \right]
\]

(A1-8-23 a, b)

Substituting for \( \sigma_{01} \) and \( \sigma_{02} \) in equation (A1-8-18)

\[
G_{12}(2d) = \frac{\varepsilon \kappa}{2} \left[ 2\psi_{01}\psi_{02} \tanh(2\kappa d) - \left( \psi_{01}^2 + \psi_{02}^2 \right) \coth(2\kappa d) \right]
\]

(A1-8-24)

It follows from equation (A1-8-24) that as separation of the two surfaces becomes large,

\[
G_{12}(\infty) = -\frac{\varepsilon \kappa}{2} \left[ \left( \psi_{01}^2 + \psi_{02}^2 \right) \coth(2\kappa d) - 1 \right]
\]

(A1-8-25)

Thus, the total potential energy density from equation (A1-8-19) may be written as

\[
V = \frac{\varepsilon \kappa}{2} \left[ 2\psi_{01}\psi_{02} \tanh(2\kappa d) - \left( \psi_{01}^2 + \psi_{02}^2 \right) \coth(2\kappa d) - 1 \right]
\]

(A1-8-26)

Equation (A1-8-26) express the potential energy density of interaction of two infinite flat surfaces as a function of the surface electric potential of each surface, and the separation of the surfaces, \( 2d \).

**Interaction Between Spherical Double Layers Due to Electric Double Layer**

The interaction between double layers on spherical particles may be assumed to be consist of contributions from infinitesimally small parallel rings each of which can be considered as a flat plate. Geometrical construction used in the calculation of the interaction between two dissimilar spherical particles are radii of \( a_1 \) and \( a_2 \) and separation of \( H_0 \). The total interaction energy between two spherical double layers is then given by

\[
U = \int_0^\infty V(H(h))2\pi h \cdot dh
\]

(A1-8-27)

where \( V \) is defined by equation (A1-8-26), \( h \) is the radius of the ring and \( H \) is separation of two rings as a function of \( h \). \( H \) can be written in terms of \( h \) as,

\[
H - H_0 = a_1 + a_2 - \sqrt{a_1^2 - h^2} - \sqrt{a_2^2 - h^2}
\]

(A1-8-28)

Differentiation of equation (A1-8-28) yields
\[
dH = \left(\frac{1}{a_1 \sqrt{(1-h^2/a_1^2)}} + \frac{1}{a_2 \sqrt{(1-h^2/a_2^2)}}\right)hdh \quad (A1-8-29)
\]

To derive \( U \) as a function of separation \( H_0 \), substitute equation \((A1-8-29)\) into \((A1-8-27)\) and we derived

\[
U = \int_{H_0}^{\infty} V(H) 2\pi \left(\frac{a_1 a_2 \sqrt{(1-h^2/a_1^2)(1-h^2/a_2^2)}}{a_1 \sqrt{(1-h^2/a_1^2)}+a_2 \sqrt{(1-h^2/a_2^2)}}\right) \cdot dH \quad (A1-8-30)
\]

To further simplify equation \((A1-8-30)\), we note that \( V(H) \) decays rapidly as \( H \) increases. When separation of \( H \) is larger than 100nm, \( V \) is \( 10^{5-6} \) smaller than EDL potential at particle surface. If particle size is in micron scale, we assume that the effective range of \( V \) is very small such that \( h \ll a_1 \) and \( h \ll a_2 \). Therefore, equation \((A1-8-30)\) reduces to

\[
U = \frac{2\pi a_1 a_2}{a_1 + a_2} \int_{H_0}^{\infty} V(H) \cdot dH \quad (A1-8-31)
\]

Substituting equation \((A1-8-26)\) into equation \((A1-8-31)\), the integral can be evaluated analytically giving

\[
U = \frac{\pi a_1 a_2 \left(\psi_{01}^2 + \psi_{02}^2\right)}{a_1 + a_2} \left[\frac{2\psi_{01}\psi_{02}}{\psi_{01}^2 + \psi_{02}^2} \ln \left(\frac{1 + \exp(-\kappa H_0)}{1 - \exp(-\kappa H_0)}\right) + \ln(1 - \exp(-2\kappa H_0))\right] \quad (A1-8-32)
\]

Interaction between a sphere and an infinite flat surface can be simply derived as,

\[
U = \frac{\pi a_1 \left(\psi_{01}^2 + \psi_{02}^2\right)}{\psi_{01}^2 + \psi_{02}^2} \left[\frac{2\psi_{01}\psi_{02}}{\psi_{01}^2 + \psi_{02}^2} \ln \left(\frac{1 + \exp(-\kappa H_0)}{1 - \exp(-\kappa H_0)}\right) + \ln(1 - \exp(-2\kappa H_0))\right] \quad (A1-8-33)
\]

Note that this relationship holds based on two important assumptions: (i) surface electric potential is low, less than 25 mV, (ii) particle size is large compared to double layer effective “thickness”.

\[
\int_{H_0}^{\infty} V(H) 2\pi \left(\frac{a_1 a_2 \sqrt{(1-h^2/a_1^2)(1-h^2/a_2^2)}}{a_1 \sqrt{(1-h^2/a_1^2)}+a_2 \sqrt{(1-h^2/a_2^2)}}\right) \cdot dH \quad (A1-8-30)
\]
APPENDIX 2. PYTHON PROGRAM FOR BACTERIAL BACKBONE DETECTION

1. ""
2. Step 1. set current working directory and prepare image for next step
3. ""
4.
5. import os
6. import scipy.misc
7.
8. from skimage import io
9. from skimage import restoration
10.
11. '''set current working directory, change according to your setting'''
    os.chdir("C:\Users\sun.j\Desktop\python cell detection")
12. '''read the image to be processed, change the image name accordingly'''
    grey_original_initial = io.imread('Tv23.jpg',as_grey=True)
    grey_original_final = io.imread('Tv25.jpg',as_grey=True)
13. '''Image Restoration'''
    grey_restored_initial = restoration.denoise_nl_means(grey_original_initial)
    grey_restored_final = restoration.denoise_nl_means(grey_original_final)
14.
15. scipy.misc.imsave('grey_restored_initial.jpg', grey_restored_initial)
16. scipy.misc.imsave('grey_restored_final.jpg', grey_restored_final)
17.
18. ""
20. Step 2. Bacterial detection and identify backbone
21. ""
22. from skimage.filter import threshold_otsu
from skimage import io
from skimage import measure
from skimage import morphology
from math import pi
from xlwt import Workbook

'''set current working directory'''
os.chdir("C:\Users\sun.j\Desktop\python cell detection")

'''read the image to be processed'''
image = io.imread('grey_restored_final.jpg')

'''set parameters'''
threshold = threshold_otsu(image)*1.3
w = 8
w1 = int(round(w/1.414))

'''define a empty ndarray and dict for each bacteria'''
binary = np.zeros(image.shape)
cell_dict = {}

'''bacterial backbone identification'''
for i in range(w,image.shape[0]-w):
    for j in range(w,image.shape[1]-w):
        if image[i,j]>threshold:
            if int(image[i,j]==np.amax(image[i-w:i+w,j-j+w]))
                + int(image[i,j]==np.amax(np.diag(image[i-w1:i+w1,j-w1:j+w1])))
                + int(image[i,j]==np.amax(np.diag(np.fliplr(image[i-w1:i+w1,j-w1:j+w1]))))>1:
                binary[i,j]=1
            else:
                binary[i,j]=0
        else:
            binary[i,j]=0

'''Refine broken backbone'''
kernel1 = np.ones((8,8),np.uint8)
kernel2 = np.ones((4,4),np.uint8)
kernel3 = np.ones((3,1),np.uint8)
closing = cv2.morphologyEx(binary,cv2.MORPH_CLOSE, kernel1,iterations=3)
dilation = cv2.dilate(binary,kernel2,iterations=1)
dilation1= cv2.dilate(dilation,kernel3,iterations=1)
scipy.misc.imsave('dilation_initial.jpg', dilation)

bact, label = measure.label(dilation, background=0, return_num=True, connectivity=2)

bact_dict = {}
i = 0

for m in range(1, label+1):
    Max_x = []
    Max_y = []
    Min_x = []
    Min_y = []
    l = -1
    bact_grey = np.multiply(bact==m, image)
    bact_binary = np.multiply(bact==m, dilation)
    area = np.sum(bact_binary)
    perimeter = measure.perimeter(bact_binary, True)
    shape_factor = area*4*pi/perimeter**2

    x_min = np.amin(np.nonzero(bact==m)[1])
    x_max = np.amax(np.nonzero(bact==m)[1])
    y_min = np.amin(np.nonzero(bact==m)[0])
    y_max = np.amax(np.nonzero(bact==m)[0])

    if area>20 and shape_factor<1 or shape_factor==1:
        i+= 1
        midline = morphology.medial_axis(bact_binary)
        if np.amax(np.nonzero(midline)[1])-np.amin(np.nonzero(midline)[1])>np.amax(np.nonzero(midline)[0])-np.amin(np.nonzero(midline)[0]):
            for k in np.nonzero(midline)[1]:
                l+= 1
                Max_x.append(np.nonzero(midline)[1][l])
                Max_y.append(np.nonzero(midline)[0][l])
            if k == np.amax(np.nonzero(midline)[1]):
                Min_x.append(np.nonzero(midline)[1][l])
        else:
            for k in np.nonzero(midline)[0]:
                l+= 1
                Min_y.append(np.nonzero(midline)[0][l])
                Max_y.append(np.nonzero(midline)[1][l])
Min_y.append(np.nonzero(midline)[0][l])

angle = math.degrees(math.atan(abs((np.mean(Max_y) - np.mean(Min_y)) / (np.mean(Max_x) - np.mean(Min_x)))))

else:
    for k in np.nonzero(midline)[0]:
        l+= 1
        if k == np.amax(np.nonzero(midline)[0]):
            Max_x.append(np.nonzero(midline)[1][l])
            Max_y.append(np.nonzero(midline)[0][l])
            angle = math.degrees(math.atan(abs((np.mean(Max_y) - np.mean(Min_y)) / (np.mean(Max_x) - np.mean(Min_x)))))
        if k == np.amin(np.nonzero(midline)[0]):
            Min_x.append(np.nonzero(midline)[1][l])
            Min_y.append(np.nonzero(midline)[0][l])

bact_dict[i] = [angle, shape_factor]

'''
save data in xls file'''
wbf = Workbook()
sheet1 = wbf.add_sheet('sheet 1')

for j in bact_dict.keys():
    sheet1.write(j-1,0,bact_dict.keys()[j-1])
    sheet1.write(j-1,1,bact_dict.values()[j-1][0])
    sheet1.write(j-1,2,bact_dict.values()[j-1][1])

wb.save('results.xls')

"""Step 3. refine the broken backbone"""

import os
import cv2
import numpy as np
import scipy.misc

from skimage import io
'''set current working directory'''

```python
os.chdir("C:\Users\sun.j\Desktop\python cell detection")
```

'''read the image to be processed'''

```python
binary = io.imread('binary_initial.jpg')
```

```python
kernel1 = np.ones((8,8),np.uint8)
kernlel2 = np.ones((4,4),np.uint8)
kernlel3 = np.ones((3,1),np.uint8)
closing = cv2.morphologyEx(binary,cv2.MORPH_CLOSE, kernel1,iterations=3)
dilation = cv2.dilate(binary,kernel2,iterations=1)
dilation1 = cv2.dilate(dilation,kernel3,iterations=1)
```

```python
scipy.misc.imsave('closing_initial.jpg', closing)
```

```python
scipy.misc.imsave('dilation_initial.jpg', dilation)
```
APPENDIX 3. DERIVATION OF FINITE DIFFERENCE SCHEMES FOR CONVECTION-DIFFUSION EQUATION AND MATLAB CODE

This appendix only focuses on the derivation of scheme for FDM and special mesh technique used in Elimelech’s work (Elimelech, 1994). The goal is to reproduce his result and apply it to our model. The convection-diffusion equation (7-18) can be written in spherical coordinates. The spherical coordinate is shown in the Figure A3.1. Therefore, the convection-diffusion equation can be reduced to

$$a_1(H, \varphi) \frac{\partial^2 C^*}{\partial H^2} + a_2(H, \varphi) \frac{\partial C^*}{\partial H} + a_3(H, \varphi) C^* = \frac{\partial C^*}{\partial \varphi}$$  \hspace{1cm} (A3-1)

Where $C^*$ is the dimensionless bacterial concentration as $C/C_0$. Detailed derivation of equation (A3-1) can be found from Elimelech’s work (Elimelech, 1994).

Figure A3.1 Spherical coordinate of a single collector. $H$ indicates the separated distance from bacteria to the collector surface. $\varphi$ indicates the location of collector surface.

The space was meshed in two dimensions of perpendicular coordinate, $H$ and tangential coordinate, $\varphi$, as shown in Figure A3.2. With the central different scheme on nonuniform step size, the first and second derivatives of equation (A3-1) are approximated by

$$\frac{\partial C^*}{\partial H} = \frac{q_k}{p_k + q_k} \frac{C^*_{k+1} - C^*_k}{p_k} + \frac{p_k}{p_k + q_k} \frac{C^*_k - C^*_{k-1}}{q_k}$$  \hspace{1cm} (A3-2)
\[
\frac{\partial^2 C^*}{\partial H^2} = \frac{2}{p_k + q_k} \left( \frac{C^*_{k+1} - C^*_k}{p_k} - \frac{C^*_k - C^*_{k-1}}{q_k} \right)
\]

(A3-3)

\[
\frac{\partial C^*}{\partial \varphi} = \frac{y_i}{x_i + y_i} \frac{C^*_{k+M} - C^*_k}{x_i} + \frac{x_i}{x_i + y_i} \frac{C^*_k - C^*_{k-M}}{y_i}
\]

(A3-4)

Variables \( p_k, q_k, x_k, \) and \( y_k \) are defined as

\[
p_k = H_{k+1} - H_k
\]

\[
q_k = H_k - H_{k-1}
\]

\[
x_i = \varphi_{i+1} - \varphi_i
\]

\[
y_i = \varphi_{i+1} - \varphi_i
\]

where \( k \in [1, M] \) and \( i \in [1, N] \).

Figure A3.2 2D special mesh on collector surface. \( H \) is perpendicular coordinate and \( \varphi \) is tangential coordinate. \( k \) and \( i \) are the indices of space variable \( H \) and \( \varphi \). \( M+2 \) is the number of nodes in \( H \) direction and \( N+2 \) is the number of nodes in \( \varphi \) direction. \( k = 0 \) represent the collector surface.

Substitute equations (A3-4) to (A3-6) into governing equation (A3-1), we derived
\[
\begin{align*}
& a_1(H, \varphi) \cdot \frac{2}{p_k + q_k} \left( C_{k+1}^* - C_k^* \right) + \frac{C_k^* - C_{k-1}^*}{q_k} + a_2(H, \varphi) \\
& + a_3(H, \varphi) C_k^* \\
& = \frac{y_i}{x_i + y_i} \frac{C_k^*}{x_i} \\
& + \frac{x_i}{x_i + y_i} \frac{C_k^* - C_{k-M}^*}{y_i} - \frac{C_k^*}{(x_i + y_i)y_i} C_{k-M}^* \\
& + \left[ a_1(H, \varphi) \cdot \frac{2}{(p_k + q_k)q_k} - a_2(H, \varphi) \right] - \frac{p_k}{(p_k + q_k)q_k} \left[ C_{k-1}^* \right] \\
& + \frac{1}{p_k + q_k} \left( \frac{q_k}{p_k} \right) + a_3(H, \theta) + \frac{1}{x_i + y_i} \\
& \cdot \left( \frac{y_i}{x_i} - \frac{x_i}{y_i} \right) C_k^* \\
& + \left[ a_1(H, \varphi) \cdot \frac{2}{(p_k + q_k)p_k} + a_2(H, \varphi) \right] \\
& \cdot \frac{q_k}{(p_k + q_k)p_k} \left[ C_{k+1}^* \right] + \left[ - \frac{y_i}{(x_i + y_i)x_i} \right] C_{k+M}^* = 0 \\
\end{align*}
\]

(A3-5)

Simplify equation (A3-5) as

\[
Q_4 C_{k-M}^* + Q_1 C_{k-1}^* + (Q_2 - Q_4 - Q_5) C_k^* + Q_3 C_{k+1}^* + Q_5 C_{k+M}^* = 0
\]

(A3-6)

Boundary condition:

- \( \varphi = 0 \) and \( \varphi = \pi \), \( a_1(H, \varphi) \frac{\partial^2 C^*}{\partial H^2} + a_2(H, \varphi) \frac{\partial C^*}{\partial H} + a_3(H, \varphi) C^* = 0 \)
  \( k = 2: (M-1) \) and \( k = (N-1)^*M+2 \) : (N*M-1)  
  \( Q_1 C_{k-1}^* + Q_2 C_k^* + Q_3 C_{k+1}^* = 0 \)

- \( H = \delta, C^* = 0 \), \( k = (n-1)^*M+1 \), \( l < i < N \)  
  \( Q_4 C_{k-M}^* + (Q_2 - Q_4 - Q_5) C_k^* + Q_3 C_{k+1}^* + Q_5 C_{k+M}^* = 0 \)

- \( H = b, C^* = 1 \), \( k = i^*M \), \( l < i < N \)  
  \( Q_4 C_{k-M}^* + Q_1 C_{k-1}^* + (Q_2 - Q_4 - Q_5) C_k^* + Q_5 C_{k+M}^* = -Q_3 \)
Here $b$ is the outer boundary and \textit{delta} is inner boundary in $H$. $b$ is set according the porous space in the porous media and bacterial concentration at this distance from collector surface is equal to bulk concentration. $\delta$ is the first point in $H$.

Therefore,

- $i=1, k=1$ \quad $Q_2 C_k^* + Q_3 C_{k+1}^* = 0$
- $i=1, k=M$ \quad $Q_1 C_{k-1}^* + Q_2 C_k^* = -Q_3$
- $i=N, k=1$ \quad $Q_2 C_k^* + Q_3 C_{k+1}^* = 0$
- $i=N, k=M$ \quad $Q_1 C_{k-1}^* + Q_2 C_k^* = -Q_3$

A nonuniform mesh technique was applied here according to Elimelech (Elimelech, 1994). An exponentially decreasing step size (decreasing toward the surface of the collector and inflow direction of $\varphi=0$) has been calculated from

$$H_k = \delta + (b - \delta) \frac{\exp\left(\frac{k \beta_1}{M}\right) - 1}{\exp(\beta_1) - 1} \quad (A3-7)$$

$$\theta_i = \frac{\pi}{1e6} + (\pi - \frac{\pi}{1e6}) \frac{\exp\left(\frac{i \beta_2}{N}\right) - 1}{\exp(\beta_2) - 1} \quad (A3-8)$$

$M$ and $N$ are number of steps in two directions. $\beta_1$ and $\beta_2$ are positive numbers adjusting the rate of change of the step size.
MATLAB CODE

%Nonuniform mesh_ inner region
clearvars
syms H theta

%Constant parameters%
C0 = 1;
k = 1.3805e-23;
T = 298;
mu = 8.9e-4;                          %viscosity of water
g = 9.8;                              %gravity constant
lamda = 0.1e-6;                       %characteristic wave length
    of the dielectric
A = 5e-20;                            %Hamaker constant
e0 = 8.854*10^-12;                    %vacuum permittivity
er = 80.1;                            %relative dielectric
    permittivity of water
e = 1.602176565e-19;                 %culum of a electron
Na = 6.02214129e23;                  %Avogadro number

%adjustable parameters
ep = 0.38;                            %porosity
Is = 100e-5;                          %ionic strength
zeta_p = 20e-3;                       %zeta potential of particle
zeta_c = -20e-3;                      %zeta potential of collector
ap = 0.2e-6;                          %particle radius
ac = 50e-6;                           %collector radius
U = 1e-5;                            %approach velocity, m/s
rho_water = 1000;                     %density of water
rho_solution = 1000;                  %density of solution
delta = 2e-5;                         %starting position of inner
    region (smaller than the
    size of an atom)
beta = 5;                             %a positive number adjusting
    the rate of change of the
    step size

b = ac*(1-ep)^(1/3)/ap;                  %dimensionless
    thickness of shell of
    fluid envelope
    according to Happel's
    model
L1 = 0.01*b;                           %dimensionless length
    of inner region
drho = rho_solution-rho_water;        %density difference
Fg = 4/3*pi*ap^3*drho*g;               %dimensionless gravity
K = (2*e^2*Is*Na/(e0*er*k*T))^0.5;     %inverse of Debye
    length
\[ D_{\text{inf}} = \frac{kT}{6\pi\mu a_p}; \quad \text{diffusion coefficient in an infinite medium}\]

\[ N_{\text{pe}} = \frac{2Ua_p}{D_{\text{inf}}}; \quad \text{particle Peclet number}\]

%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
% Mesh
% Define mesh in H direction
M_in = 500; \quad \%Number of elements in inner region
M_out = 2500; \quad \%Number of elements in outer region
HH = zeros(1,M_in+M_out+1);
HH_out = linspace(L1+delta+(b-L1-delta)/M_out,b,M_out);
dH_out = (b-L1-delta)/M_out;
I = 0:M_in;
HH(I:M_in+1) = delta+(L1-delta)*(exp(I.*beta/M_in)-1).
\%inner region nonuniform mesh
HH_out = linspace(L1+delta+(b-L1-delta)/M_out,b,M_out);
HH(M_in+2:M_in+M_out+1) = HH_out; \quad \%outer region uniform mesh

% Define mesh in theta direction
N = 101;
Theta = linspace(0,\pi-\pi/1e6,N); \%Number of nodes in theta direction
dtheta = Theta(N)/(N-1);
C = zeros(M_in+M_out+1,N); \%concentration distribution

% Define tridiagonal matrix coefficient b d e f%
B1 = ones(M_in+M_out+1,N);
D1 = ones(M_in+M_out+1,N);
E1 = ones(M_in+M_out+1,N);
F = zeros(M_in+M_out+1,N);

% Boundary conditions%
D1(1,:) = 1; \quad \%C=0 \@ H=delta
E1(1,:) = 0; \quad \%C=0 \@ H=delta
F(M_in+M_out+1,:) = 1; \quad \%C=1 \@ H=inf
B1(M_in+M_out+1,:) = 0; \quad \%C=1 \@ H=inf
D1(M_in+M_out+1,:) = 1; \quad \%C=1 \@ H=inf

% solving for initial condition %
% solving for initial condition %
theta=0;
for i=2:M_in+M_out
    \% inner region
    if i <=M_in+1
        H = HH(i);
        \%---------------------------------------
    \end{if}
%dependent variables
%-------------------------------------
Nr = ap/(ac+H*ap);

p = HH(i+1) - HH(i);
g = HH(i) - HH(i-1);
if H < 0.0001
  f1 = H/(1+H);
  f2 = 3.2295;
  f3 = 0.7431/(0.6376-0.2*log(H));
  f4 = -2*log(H);
  df1dH = 1/(H+1) - H/(H+1)^2;
  df2dH = 0;
%Nondimentionalized flow field
Vr = -cos(theta)*((ac/(ac+H*ap))^3/2-3*ac/(2*(ac+H*ap))+1);
Vtheta = sin(theta)*(-(ac/(ac+H*ap))^3/4-3*ac/(4*(ac+H*ap)))+1);
dVrdH = -
cos(theta)*((6*ac*ap)/(2*ac+2*H*ap)^2-3*ac^3*ap)/(2*(ac+H*ap)^4));
%Nondimentionalized DLVO potential and DLVO force
phi_non = -
  A/(6*H*(1+14*H*ap/lamda)*k*T)+(pi*e0*er*ap/(k*T))*(2*zeta_p*zeta_c*log(1+exp(-
  K*H*ap))/((1-exp(- K*H*ap)))+1)*((zeta_p^2+zeta_c^2)*log(1-
  exp(-2*K*H*ap)));
dphidH = A/(6*H^2*T*k*((14*H*ap)/lamda + 1))
  - (ap*e0*er*pi/(k*T))*(2*zeta_p*zeta_c^2*log(1+exp(-
  K*H*ap)-1)))/(exp(-
  2*K*H*ap)-1) - (2*zeta_c*zeta_p*exp(-
  H*K*ap)-1)*((K*ap*exp(-H*K*ap))/((1-exp(-
  H*K*ap)-1) - (K*ap*exp(-H*K*ap)+1))/(exp(-
  H*K*ap)+1)) + (7*A*ap)/(3*H*T*k*lamda*((14*H*ap)/lamda + 1)^2);
d2phidH2 =
  (ap*e0*er*pi/(k*T))*(4*K^2*ap^2*exp(-
  2*K*H*ap)*(-zeta_p^2+zeta_p^2))/((exp(-
  2*K*H*ap)-1) - (4*K^2*ap^2*exp(-
  4*H*K*ap)))*((zeta_c^2+zeta_p^2))/((exp(-
  2*K*H*ap)-1)^2 +
  (2*zeta_c*zeta_p*exp(-H*K*ap) - 1)*((K^2*ap^2*exp(-H*K*ap))/((exp(-
  H*K*ap)-1) - (K^2*ap^2*exp(-
  H*K*ap)+1))/(exp(-H*K*ap)+1)) + (7*A*ap)/(3*H*T*k*lamda*{(14*H*ap)/lamda + 1)^2);
\[
2*H*K*ap)*((exp(-H*K*ap) - 1)^(3))/((exp(-H*K*ap) + 1) + (2*K*ap*zeta_c*zeta_p*exp(-H*K*ap))*(K*ap*exp(-H*K*ap))/((exp(-H*K*ap) - 1) - (K*ap*exp(-H*K*ap))*(exp(-H*K*ap) + 1)))/((exp(-H*K*ap) - 1)^2))/((exp(-H*K*ap) + 1)^2) - (2*K*ap*zeta_c*zeta_p*exp(-H*K*ap))*(exp(-H*K*ap) - 1)*((K*ap*exp(-H*K*ap))/((exp(-H*K*ap) - 1) - (K*ap*exp(-H*K*ap))*(exp(-H*K*ap) + 1)))/((exp(-H*K*ap) - 1)^2))/((exp(-H*K*ap) + 1)^2) - A/(3*H^3*T*k*((14*H*ap)/lamda + 1)) - (14*A*ap)/(3*H^2*T*k*lamda*((14*H*ap)/lamda + 1)^2) - (196*A*ap^2)/(3*H*T*k*lamda^2*((14*H*ap)/lamda + 1)^3);
\]

%coefficients of modified governing equation%
a1 = f1; \%this is for theta>0
a2 = -(df1dH-2*f1*Nr+1/2*f1*f2*Npe*Vr-f1*(dphidH+Fg*cos(theta)));
a3 = -(d2phidH2*f1+2*f4*Fg*Nr*cos(theta) - (df1dH+2*f1*Nr)*(dphidH+Fg*cos(theta)) + 1/2*Npe*(dVrdH*f1*f2+df2dH*f1+df1dH* f2+2*f1*f2*Nr)*Vr+2*f3*Nr*(-(ac/(ac+H*ap))^3/4 - 3*ac/(4*(ac+H*ap))�))/1));

%coefficients of discretized PDE
B1(i,1) = (2*a1-a2*p)/((p+q)*q);
D1(i,1) = -2*a1*(1/p+1/q)/(p+q)-a2*(q/p- p/q)/(p+q)+a3;
E1(i,1) = (2*a1+q*a2)/((p+q)*p);
else \%H>0,0001
%universal function of hydrodynamic interaction
f1 = H/(1+H); f2 = (1-17.69/(37*(H+1.416)))/(2.85); f3 = 1-1.668/(2.396*(H+1)^2); f4 = 1-6.259/(2.256*(H+2)^2);
df1dH = 1/(H + 1) - H/(H + 1)^2; df2dH = -((121017*exp(-1391*H)/1000))/50000 - (605241*exp(-819*H)/25000))/12500000;
%Nondimentionalized flow field
Vr = -cos(theta)*((ac/(ac+H*ap))^3/2- 3*ac/(2*(ac+H*ap))+1); Vtheta = sin(theta)*(-((ac/(ac+H*ap))^3/4- 3*ac/(4*(ac+H*ap))+1);
\[ dV_{rdH} = - \cos(\theta) \frac{(6ac*ap)}{(2ac+2H*ap)^2 - (3ac^3*3ap)^2/(2*(ac+H*ap)^4)}; \]

\%Non-dimentionalized DLVO potential and DLVO force\%
\[
\phi_{non} = - A/(6H^2*T*k*((14*H*ap)/lamda + 1)) - (ap*e0*er*pi/(k*T))*((2*K*ap*exp(-2*H*K*ap))^2 + (2*zeta_c*zeta_p*exp(-H*K*ap) - 1))*((K*ap*exp(-H*K*ap)))/(exp(-H*K*ap) - 1) - 1) + (2*zeta_c*zeta_p*exp(-H*K*ap))/(exp(-H*K*ap) - 1) + (7*A*ap)/(3*H*T*k*lamda)*((14*H*ap)/lamda + 1)^2; \]

\[ d\phi/dH = A/(6H^2*T*k*((14*H*ap)/lamda + 1)) - (ap*e0*er*pi/(k*T))*((2*K*ap*exp(-2*H*K*ap))^2 + (2*zeta_c*zeta_p*exp(-H*K*ap) - 1))*((K*ap*exp(-H*K*ap)))/(exp(-H*K*ap) - 1) + (7*A*ap)/(3*H*T*k*lamda)*((14*H*ap)/lamda + 1)^2; \]

\[ d^2\phi/dH^2 = (ap*e0*er*pi/(k*T))*((4*K^2*ap^2*exp(-2*H*K*ap))^2 + (2*H*K*ap))^2 - (K*ap*exp(-H*K*ap) + 1))/(exp(-H*K*ap) - 1) + (2*K*ap*zeta_c*zeta_p*exp(-H*K*ap))/(exp(-H*K*ap) - 1) + (7*A*ap)/(3*H*T*k*lamda)*((14*H*ap)/lamda + 1)^2; \]

\%coefficients of modified governing equation\%
\[ a1 = f1; \]
\[ a_2 = -(df_{1dH} - 2f_1 N_r + f_1 f_2 N_{pe} V_r - f_1 (d\phi_{1dH} F_g \cos(\theta))) \]
\[ a_3 = -(d^2 \phi_{1dH}^2 f_1 + 2f_4 F_g N_r \cos(\theta)) \] (1)
\[ \frac{df_1dH}{f_1} + 2f_1 N_r (d\phi_{1dH} + F_g \cos(\theta)) + \frac{1}{2} N_{pe} ((dV_r dH f_1 + df_{1dH} f_2 + 2f_1 f_2 N_r) V_r + 2f_3 N_r (-ac/(ac+H*ap))^{3/4} - 3ac/(4*(ac+H*ap))^{1/3})) ; \]

\% coefficients of discretized PDE
\[ B_1(i,1) = (2a_1 - a_2 p)/(p+q)q ; \]
\[ D_1(i,1) = -2a_1 (1/p + 1/q)/(p+q) - a_2 (q/p - p/q)/(p+q) + a_3 ; \]
\[ E_1(i,1) = (2a_1 + q a_2)/(p+q) p ; \]

end

\% Outer region
\%-------------------------------------------------------
\% Outer region
\%-------------------------------------------------------

\[ H = HH_{out}(i-M_{in}-1) ; \]
\[ N_r = ap/(ac+H*ap) ; \]
\[ f_1 = H/(1+H) ; \]
\[ f_2 = (1-17.69/(37*(H+1.416)))^{(-2.85)} ; \]
\[ f_3 = 1-1.668/(2.396*(H+1)^5) ; \]
\[ f_4 = 1-6.259/(2.256*(H+2)^2) ; \]
\[ df_{1dH} = 1/(H + 1) - H/(H + 1) ; \]
\[ df_{2dH} = -(121017*exp(-1391*H)/10000 - 605241*exp(-819*H)/25000)/12500000 ; \]

\% Nondimensionalized flow field
\[ V_r = -\cos(\theta) *((ac/(ac+H*ap))^{3/2} - 3ac/(2*(ac+H*ap)) + 1) ; \]
\[ V_{theta} = \sin(\theta) *((ac/(ac+H*ap))^{3/4} - 3ac/(4*(ac+H*ap)) + 1) ; \]
\[ dV_r dH = \cos(\theta) *((6ac*ap)/(2ac+2H*ap)^2 - (3ac^3*ap)/(2(ac+H*ap))^{1/2}) ; \]

\% Nondimensionalized DLVO potential and DLVO force
\[ \phi_{non} = \frac{A}{(6H^2*T*K*ap/\lambda) + (pi*e0*er*ap)/(k*T)} (2*zeta_p*zeta_c^2) \log((1+exp(-K*H*ap))/\exp(-K*H*ap)) + (zeta_p^2+2+zeta_c^2) \log(1-\exp(-2*K*H*ap)) ; \]
\[ d\phi_{1dH} = \frac{A}{6H^2*T*K*ap/\lambda} (2*zeta_p*zeta_c^2) \log((1+exp(-K*H*ap))/\exp(-K*H*ap)) + (zeta_p^2+2+zeta_c^2) \log(1-\exp(-2*K*H*ap)) ; \]
\[
\frac{\left(\frac{H^{*}K^{*}a}{p}+1\right)}{\left(\exp\left(-H^{*}K^{*}a\right)+1\right)^{2}} = \frac{\left(\frac{H^{*}K^{*}a}{p}+1\right)+\left(\frac{7}{H^{*}K^{*}a}\right)}{\left(\frac{1}{H^{*}T^{*}k^{*}\lambda}\right)^{2}} = \left(\frac{1}{H^{*}K^{*}a}\right)^{2} + \left(\frac{1}{H^{*}T^{*}k^{*}\lambda}\right)^{2}.
\]

\[
d2\phi/2 = \frac{\left(\frac{e_{0}e_{r}\pi}{k^{*}T}\right)}{\left(\frac{1}{H^{*}K^{*}a}\right)^{2}} = \frac{\left(\frac{1}{H^{*}K^{*}a}\right)^{2} + \left(\frac{1}{H^{*}T^{*}k^{*}\lambda}\right)^{2}}{\left(\frac{1}{H^{*}K^{*}a}\right)^{2} + \left(\frac{1}{H^{*}T^{*}k^{*}\lambda}\right)^{2}}.
\]

% coefficients of modified governing equation
\[
a_{1} = f_{1};
a_{2} = -\left(\frac{df_{1}dH}{2} + f_{1} + f_{2} + N_{pe}V_{r}\right) = \left(\frac{df_{1}dH}{2} + f_{1} + f_{2} + N_{pe}V_{r}\right) = \left(\frac{df_{1}dH}{2} + f_{1} + f_{2} + N_{pe}V_{r}\right) = \left(\frac{df_{1}dH}{2} + f_{1} + f_{2} + N_{pe}V_{r}\right).
\]

% coefficients of discretized PDE
\[
B_{1}(i,1) = a_{1}/dH_{out}^{2};
D_{1}(i,1) = -2*a_{1}/dH_{out}^{2} - a_{2}/dH_{out} + a_{3};
E_{1}(i,1) = a_{1}/dH_{out}^{2} + a_{2}/dH_{out};
\]

end
C(:,1) = TDMAsolver(B1(:,1),D1(:,1),E1(:,1),F(:,1));
figure(1)
plot(HH,C(:,1))
hold on
tic;

for j = 2:N-1
    theta_mid = Theta(j)-dtheta/2;
    theta = theta_mid;
    for i=2:M_in+M_out
        if i <= M_in+1
            H = HH(i);
        end
    end
    theta = theta_mid;
end

% inner region
if i <= M_in+1
    H = HH(i);
end

% dependent variables
%-------------------------------------
Nr = ap/(ac+H*ap);
p = HH(i+1)-HH(i);
q = HH(i)-HH(i-1);
if H < 0.0001
    f1 = H/(1+H);
    f2 = 3.2295;
    f3 = 0.7431/(0.6376-0.2*log(H));
    f4 = -2*log(H);
    df1dH = 1/(H + 1) - H/(H + 1)^2;
    df2dH = 0;
end

% Nondimensionalized flow field
Vr = -cos(theta) *((ac/(ac+H*ap))^3/2-
    3*ac/(2*(ac+H*ap))+1);
Vtheta = sin(theta) * (-ac/(ac+H*ap))^3/4-
    3*ac/(4*(ac+H*ap))+1);
dVrdH = -
    cos(theta) *((6*ac*ap)/(2*ac+2*H*ap)^2-
    (3*ac^3*ap)/(2*(ac+H*ap)^4));

% Nondimensionalized DLVO potential and
DLVO force
phi_non = -
    A/(6*H*(1+14*H*ap/lamda)*k*T)+(pi*e0*er*ap/
    (k*T))* (2*zeta_p*zeta_c* log((1+exp(-
    K*H*ap))/(1-exp(-
    K*H*ap))))+(zeta_p^2+zeta_c^2)* log(1-exp(-
    2*H*K*ap)));
dphidH = A/(6*H^2*T*k*((14*H*ap)/lamda +
    1)) - (ap*e0*er*pi/(k*T))* ((2*K*ap*exp(-
    2*H*K*ap)* (zeta_c^2 + zeta_p^2))/(exp(-
    2*H*K*ap) - 1) + (2*zeta_c*zeta_p* (exp(-
    H*K*ap)-1)*((K*ap*exp(-H*K*ap))/(exp(-
    H*K*ap)-1)-(K*ap*exp(-H*K*ap))* (exp(-
    H*K*ap)+1))/(exp(-H*K*ap)+1))^2))/(
    (7*A*ap)/(3*H*T*k*lamda*((14*
    H*ap)/lamda + 1)^2));
\[
d2\phi/dH^2 = (ap*e0*er*pi/(k*T))*((4*K^2*ap^2*exp(-2*H*K*ap) - 1) - (4*K^2*ap^2*exp(-H*K*ap)*zeta_c^2 + zeta_p^2) - 2*H*K*ap)/(exp(-2*H*K*ap) - 1)\]^2 + (2*zeta_c*zeta_p*exp(-H*K*ap) - 1)*)\exp(-H*K*ap) - (K^2*ap^2*exp(-H*K*ap) + 1))/(exp(-H*K*ap) - 1)^3)))/(exp(-H*K*ap) + 1) + (2*K*ap*zeta_c*zeta_p*exp(-H*K*ap)*exp(-H*K*ap) - 1) - (K*ap*exp(-H*K*ap) - 1)/(exp(-H*K*ap) - 1)^2)/exp(-H*K*ap) + 1) - (2*K*ap*zeta_c*zeta_p*exp(-H*K*ap) - 1)*(K*ap*exp(-H*K*ap) - 1)/(exp(-H*K*ap) - 1) - (K*ap*exp(-H*K*ap) - 1)/(exp(-H*K*ap) - 1)^2)/(exp(-H*K*ap) - 1)^2) - A/(3*H^3*T*k*((14*H*ap)/lamda + 1)) - (196*A*ap^2)/(3*H*T*k*lamda^2*(14*H*ap)/lamda + 1)^3);

%coefficients of modified governing equation%
\[a_1 = f1/(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta));\]
\[a_2 = -(df1dH-2*f1*Nr+1/2*f1*f2*Npe*Vr-f1*(dphiH+Fg*cos(theta)))/f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta));\]
\[a_3 = -d2phiH2*f1+2*f4*Fg*Nr*cos(theta) - (df1dH^2*f1*Nr)*(df1H+Fg*cos(theta)) + 1/2*Npe*(dVrdH*f1*f2 + (df2dH*f1+df1dH*f2+2*f1*f2*Nr)*Vr) + 2*f3*Nr*Vtheta*cot(theta));\]

%coefficients of discretized PDE
\[B1(i,j) = -dtheta*(a1-a2*p/2)/(p+q);\]
\[D1(i,j) = 1-dtheta*((a1*(1/q-1/p)-a2/2*(q/p-p/q))/(p+q)+a3/2);\]
\[E1(i,j) = -dtheta*(a1+a2*q/2)/(p+q));\]
\( F(i,j) = -B_1(i,j) \cdot C(i-1,j-1) + (2 - D_1(i,j)) \cdot C(i,j-1) - E_1(i,j) \cdot C(i+1,j-1); \)

\( \text{else} \quad \% \text{H} > 0.0001 \)

\( f_1 = H/(1+H); \)
\( f_2 = (1-17.69/(37*(H+1.416)))^{(-2.85)}; \)
\( f_3 = 1-1.668/(2.396*(H+1)^5); \)
\( f_4 = 1-6.259/(2.256*(H+2)^2); \)
\( d\text{f1}dH = 1/(H + 1) - H/(H + 1)^2; \)
\( d\text{f2}dH = - (121017*\exp(-1391*H)/1000))/50000 - (605241*\exp(-819*H)/25000))/12500000; \)

\%Nondimensionalized flow field
\( V_r = -\cos(\theta)(1/(ac/(ac+H*ap)))^3/2-3*ac/(2*(ac+H*ap)+1); \)
\( V_{\theta} = \sin(\theta)*(-1/(ac/(ac+H*ap)))^3/4-3*ac/(4*(ac+H*ap)+1); \)
\( dV_{rdH} = - \cos(\theta)*((6*ac*ap)/(2*ac+2*H*ap)^2-3*ac^3*ap/(2*(ac+H*ap)^4)); \)

\%Nondimensionalized DLVO potential and DLVO force
\( \phi_{\text{non}} = - A/(6*H^2*T*k*((14*H*ap)/\text{lamda} + 1)) - (\text{ap}*e0*er*ap/(k*T))*((2*ap*exp(-2*H*K*ap)+1)*((2*ap*exp(-H*K*ap)+1))/((2*ap*exp(-H*K*ap)-1)+((2*ap*exp(-H*K*ap)-1)*((2*ap*exp(-H*K*ap)))/((2*ap*exp(-H*K*ap)-1)-((2*ap*exp(-H*K*ap)-1)/((2*ap*exp(-H*K*ap)-1)^2))); \)
\( d\phi_{dH} = (ap*e0*er*ap/(k*T))*((2*ap*exp(-2*H*K*ap)+1)-(2*ap*exp(-2*H*K*ap)+1))/((2*ap*exp(-2*H*K*ap)+1)-(2*ap*exp(-2*H*K*ap)+1)/((2*ap*exp(-2*H*K*ap)+1))); \)
\( d^2\phi_{dH^2} = (ap*e0*er*ap/(k*T))*((2*ap*exp(-2*H*K*ap)+1)-(2*ap*exp(-2*H*K*ap)+1))/((2*ap*exp(-2*H*K*ap)+1)-(2*ap*exp(-2*H*K*ap)+1)/((2*ap*exp(-2*H*K*ap)+1)); \)
\[
\begin{align*}
H*K*ap - 1)^3)/(exp(-H*K*ap) + 1) + \\
(2*K*ap*zeta_c*zeta_p*exp(-
H*K*ap)/(exp(-H*K*ap) - 1) - (K*ap*exp(-H*K*ap)*exp(-
H*K*ap) + 1))/(exp(-H*K*ap) - 1)^2)/((exp(-H*K*ap) + 1)^2) - \\
A/(3*H^3*T*k*((14*H*ap)/lamda + 1)) - \\
(14*A*ap)/(3*H^2*T*k*lamda*((14*H*ap)/lamda + 1)^2) - \\
(196*A*ap^2)/(3*H*T*k*lamda^2*((14*H*ap)/lamda + 1)^3);
\end{align*}
\]

%coefficients of modified governing equation
\[
\begin{align*}
a1 &= f1/(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta)); \\
a2 &= -(df1dH-2*f1*Nr+1/2*f1*f2*Npe*Vr-
(f1*(dphidH+Fg*cos(theta)))/(1/2*f3*Npe*Nr
*Vtheta+f4*Fg*Nr*sin(theta)); \\
a3 &= -(d2phidH2*f1+2*f4*Fg*Nr*cos(theta)-
(df1dH+2*f1*Nr)*(dphidH+Fg*cos(theta))) + 1/2*Npe* (dVrdH*f1+f2+df1dH*f2+2*f1*f2*Nr)*Vr + 2*f3*Nr*Vtheta*cot(theta))/
(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta));
\end{align*}
\]

%coefficients of discretized PDE
\[
\begin{align*}
B1(i,j) &= -dtheta*(a1-a2*p/2)/((p+q)*q); \\
D1(i,j) &= 1-dtheta*((a1*(-1/q-1/p)-a2/2*(p+q)+a3/2); \\
E1(i,j) &= -dtheta*(a1+a2*q/2)/((p+q)*p); \\
F(i,j) &= -B1(i,j)*C(i-1,j-1)+(2-
D1(i,j))*C(i,j-1)-E1(i,j)*C(i+1,j-1); \\
end
else
%
%Outer region%
%
\[
\begin{align*}
H &= HH_out(i-M_in-1); \\
Nr &= ap/(ac+H*ap); \\
f1 &= H/(1+H);
\end{align*}
\]
\[ f_2 = (1-17.69/(37*(H+1.416)))^{(-2.85)}; \]
\[ f_3 = 1-1.668/(2.396*(H+1)^5); \]
\[ f_4 = 1-6.259/(2.256*(H+2)^2); \]
\[ \text{df1dH} = \frac{1}{H + 1} - \frac{H}{(H + 1)^2}; \]
\[ \text{df2dH} = - (121017*\exp((-1391*H)/1000))/50000 - (605241*\exp((-819*H)/25000))/12500000; \]
\% Nondimensionalized flow field
\[ V_r = -\cos(\theta) \times ((ac/(ac+H*ap))^3/2 - 3*ac/(2*(ac+H*ap)) + 1); \]
\[ V_{\theta} = \sin(\theta) \times ((ac/(ac+H*ap))^3/4 - 3*ac/(4*(ac+H*ap)) + 1); \]
\[ \text{dVrdH} = - \cos(\theta) \times ((6*ac*ap)/(2*ac+2*H*ap)^2 - (3*ac^3)/(2*(ac+H*ap)^4)); \]

\% Nondimensionalized DLVO potential and DLVO force
\[ \phi_{non} = - \frac{A}{(6*H^2*T*k* ((14*H*ap)/\lambda + 1)}) \times \cos(\theta) \times ((6*ac*ap)/(2*ac+2*H*ap)^2 - (3*ac^3)/(2*(ac+H*ap)^4)); \]
\[ \frac{d\phi}{dH} = \frac{A}{(6*H^2*T*k* ((14*H*ap)/\lambda + 1))}; \]
\[ \frac{d^2\phi}{dH^2} = \frac{A}{(6*H^2*T*k* ((14*H*ap)/\lambda + 1))}; \]
\[(2*K*ap*zeta_c*zeta_p*\exp(-H*K*ap)*(\exp(-H*K*ap) - 1)*((K*ap*\exp(-H*K*ap))/\exp(-H*K*ap) - 1) - (K*ap*\exp(-H*K*ap)*(\exp(-H*K*ap) + 1))/\exp(-H*K*ap) - 1)^2)/(\exp(-H*K*ap) + 1)^2) - A/(3*H^3*T*k*((14*H*ap)/\lambda + 1)) - (14*A*ap)/(3*H^2*T*k*\lambda + ((14*H*ap)/\lambda + 1)^2) - (196*A*ap^2)/(3*H*T*k*\lambda^2*(((14*H*ap)/\lambda + 1)^3));

% coefficients of modified governing equation
a1 = f1/(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta));
a2 = -(d*dH-f1*Nr+1/2*f1*f2*Npe*Vr-f1*(dphidH+Fg*cos(theta)))/(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta));
a3 = -(d2phidH2*f1+2*f4*Fg*Nr*cos(theta)-(d*dH2*f1*Nr)*(dphidH+Fg*cos(theta)))/1/2*Npe*(dVrdH*f1+f2*(dVrdH2*f1+df1dH*f2+2*f1*f2*Nr)*Vr)+2*f3*Nr*Vtheta*cot(theta)/(1/2*f3*Npe*Nr*Vtheta+f4*Fg*Nr*sin(theta)) ;

% coefficients of discretized PDE
B1(i,j) = -a1*dtheta/(2*dH_out^2)+a2*dtheta/(4*dH_out);
D1(i,j) = 1+a1*dtheta/dH_out^2-a3*dtheta/2;
E1(i,j) = -a1*dtheta/(2*dH_out^2)-a2*dtheta/(4*dH_out);
F(i,j) = -B1(i,j)*C(i-1,j-1)+(2-D1(i,j))*C(i,j-1)-E1(i,j)*C(i+1,j-1);

end
C(:,j) = TDMAsolver(B1(:,j),D1(:,j),E1(:,j),F(:,j));
end
toc;
figure(2)
surf(Theta,HH,C)
hold on
%Calculate flux at H>0
dCdH0 = (C(2,:)-C(1,:))/(HH(2)-HH(1)); %dCdH at H>0
H0 = (HH(2)-HH(1))/2;
Vr0 = -cos(Theta).*((ac/(ac+H0*ap))^3/2-3*ac/(2*(ac+H0*ap))+1);
dphidH0 = A/(6*H0^2*T*k*((14*H0*ap)/\lambda + 1)) - ap*e0*er*p*((2*K*ap*\exp(-2*H0*K*ap)*(zeta_c^2 + zeta_p^2))/(\exp(-2*H0*K*ap) - 1) +
(2*zeta_c*zeta_p*(exp(-H0*K*ap) - 1)*((K*ap*exp(-H0*K*ap) - 1) - (K*ap*exp(-H0*K*ap))*(exp(-H0*K*ap) + 1))/(exp(-H0*K*ap) - 1)^2))/(exp(-H0*K*ap) + 1)) + (7*A*ap)/(3*H0*T*k*lamda*((14*H0*ap)/lamda + 1)^2);

f1_0 = H0/(1+H0);
f2_0 = 3.2295;
J_dimensionless = -2*f1_0*dCdH0/Npe+f1_0*f2_0*Vr0.*C(2,:)/2-2*f1_0*dphidH0*C(2,:)/(2*Npe);
J0 = U*C0.*J_dimensionless;

%Calculate overall rate of particle deposition on the collector, I
I0 = 2*pi*ac^2*trapz(Theta,J0);
eta = I0/(pi*ac^2*U*C0);
VITA

Jianfeng Sun was born on December 3\textsuperscript{rd} 1983 in Weihai City, Shandong Province, P. R. China, where he lived until graduation from Weihai No. 2 Middle School in July, 2002. He then attended Shandong University and received the bachelor degree majoring in Thermal Engineering in June, 2006. After graduation, he left the engineering field and worked in Qingdao TV station as a camera operator. In 2011, he decided to return to the engineering field and went to the United States pursuing a M.S. degree in Mechanical Engineering in Northeastern University in Boston. In 2013 he continued his Ph.D. program in Prof. Kai-tak Wan’s group in Northeastern University. He was held a Graduate Research Assistantship from Department of Mechanical and Industrial Engineering in Northeastern University during the PhD study. He is a member of Adhesion Society. In December 2017, he received his PhD in Mechanical Engineering from Northeastern University.