A NUMERICAL STUDY FOR LIQUID BRIDGE BASED MICROGRIPPING AND
CONTACT ANGLE MANIPULATION BY ELECTROWETTING METHOD

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ABSTRACT

The last decade had witnessed some very outstanding research on Micro Electro Mechanical Systems (MEMS) that has vast impact on future technologies. However building a complete microsystem requires proper microassembly methods. But microassembly research is facing stiff resistance due to the presence of many dominant forces which appears due to the scaling law. Overcoming these forces has been found to be a major drawback in microgripper research. So the primary the challenge today researches face is the lack of proper manipulation schemes. Understanding the physical forces associated with micro-scale as well as devising techniques to control them is needed in order to design a proper micromanipulation scheme.

The motivation of this study is to contribute to the microassembly research by studying the forces in microscale and propose a micromanipulation scheme to control these forces. A pick and place technique using a liquid bridge based microgripper is presented in this dissertation. Studies conducted on liquid bridge based single probe gripper have shown promise in picking up an object using strong capillary force and surface tension forces as the lifting forces. But a smooth release of the object has been a challenge. We have proposed a novel manipulation scheme by using electrowetting method to control the lifting forces. By changing the electrical field imposed on the
gripper surface, one can change the contact angle of the liquid bridge and therefore can change the meniscus geometry. Change in curvature of the meniscus causes the lifting forces between the object and the gripper to change. The focus of this study is to explore the possibility of breaking a liquid bridge by increasing the contact angles for a smooth release of an object.

A theoretical study was conducted to understand the effect of contact angle manipulation on the lifting forces. Young-Laplace equation, the non linear differential equation governing the liquid interface is numerically solved for a constant liquid volume and the specified contact angles as boundary conditions. Another set of numerical solution using commercial multi-physics software CFDACE+ has been used in parallel to validate the hypothesis. Results show that for a proper choice of liquid volume the contact angle manipulation is suitable for picking up and releasing of an object.
DEDICATION

I dedicate this dissertation to my parents

Dr. Chirasukh Chandra

Mrs. Indira Chandra
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First and foremost I would like to express my sincere gratitude to my advisor Dr. Celal Batur, for providing me the opportunity to work under his supervision. Without his kind support and guidance this work would not have been possible. He has taught me how to approach a research problem from the basics, how to ask questions and be persistent to find the answers.

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CHAPTER I

INTRODUCTION

“There is plenty of room at the bottom”

Richard Feynman, 1959

On December 29, 1959 noble prize winner physicist, Dr. Richard Feynman gave a classic talk entitled “There is plenty of room at the bottom” which inspired a whole new technology. Since then active research is being pursued towards understanding the physics and chemistry in the microscale and nanoscale and the next part of the 20th century witnessed tremendous advances in micro and nanotechnology. As these technologies evolved, scientists were able to apply the knowledge in creating innovative and challenging applications for the benefit of mankind. In the last few decades, major advances in fabrication procedures for Integrated Circuits (IC) and Micro Electro Mechanical Systems (MEMS) have revealed the prospect of creating complex miniature components. Combining these components for design of a complete microsystem involves assembly of these different components; build through different fabrication procedures, on a microassembly platform. But as the scale of the object reduces the well established principles associated with the usual assembly process in the meso scale were found to be inappropriate. As a result research on microassembly became necessary for
further advancement in MEMS design. Designing suitable microgrippers and micromanipulation schemes was found to be a fundamental requirement for microassembly research. The first step in planning effective microassembly techniques is to understand the dominant forces associated in microscale and effective manipulation schemes to control those forces. The need for fundamental research on this area has been the motivation of this study and this dissertation aims at understanding the challenges associated with microgrippers design and make a contribution to microassembly research by proposing a novel microgripping technique.

1.1 Definition of The Problem

Microassembly research is in its nascent stage and there is ample scope of research for designing cost-effective devices and schemes for manipulation of small objects. For a pick and place like routine the most effective manipulation tool that can be used is a tweezer and so it has been a trend to imitate the tweezer like design in microscale to develop complicated microgrippers. Experiments on these microgrippers reported presence of some adhesion forces and the magnitudes of these forces were found to be significantly large to overcome. So when the tweezers approaches the object, the object tends to snap on to the tweezer surface due to these strong adhesion forces and then releasing the object from the gripper surface becomes a problem. So what are these adhesion forces? In microscale volume dependent gravitational force becomes negligible and surface area dependent forces like capillary force, surface tension force, Van der Waal’s force, electrostatic force emerges as the dominant forces due to scaling law.
Research has been performed to overcome these unwanted forces through surface modification, surface coating. This dissertation aims to find an alternative solution to this problem by studying the feasibility of using these unwanted adhesive forces in favor of manipulation. A comparative study in Chapter II reveals that the most dominant forces in micro scale are the capillary and surface tension forces. A liquid bridge based gripper model has been proposed here which aims to utilize the capillary and surface tension forces as the lifting force. Furthermore a novel manipulation scheme has been proposed for the first time, where the lifting forces can be manipulated by the electrowetting method. Electrowetting is a phenomenon where surface energy changes when potential is applied to the surface, resulting in change in contact angle. Also it has been noticed that the tweezer like design is based on contact between the gripper and object surface, which proves to be detrimental to delicate components as it leaves indentations and scratch marks after being released. A liquid bridge based gripping would provide a soft gripping of the object and reduce damage of delicate components. For simplicity a case study was selected where the object is considered to be a spherical object, and the gripper surface is a flat surface. The proposed gripper surface would have an electrowetting based dielectric design (EWOD). The gripper surface would have a layer of dielectric coating on top of an electrode. A ground wire would be passed through the dielectric layer and another thin layer of hydrophobic coating would be applied on top of the surface. The initial surface would be hydrophobic with a contact angle $\sim 120^\circ$. Application of voltage would reduce the contact angle to contact angle $\sim 70^\circ$.

There would be four manipulations steps in the proposed scheme as demonstrated by Figure 1.1
1. Approach

2. Pickup

3. Move

4. Release

Approach

- Initial gripper surface should be hydrophobic.
- Application of voltage would change the surface to hydrophilic.
- A small liquid droplet would be placed on the surface
- Gripper would be positioned above the object
- Gripper would approach the object vertically from the above

Pickup

- As the gripper approaches the object liquid droplet would touch the object
- Liquid bridge would be formed between the object and the gripper surface
- Capillary and surface tension forces would act as lifting force for the object
- Object would be picked up

Move

- The gripper moves to the assembly location
Figure 1.1: Schematic representation of the micromanipulation scheme (a) approach, (b) pickup, (c) move (d) release.

**Release**

- Gripper would be positioned above the release location
• As the voltage is released, contact angle would increase, lifting forces would decrease

• Object would be released when
  ▪ Lifting force is less than the weight of the object
  ▪ Liquid bridge ruptures at some critical value of the contact angle
  ▪ Liquid bridge is made unstable by slight disturbance applied to the system
  ▪ Adhesive force of the assembly platform can be used for release by moving the object close to the assembly surface to allow another liquid bridge to be formed between the object and the assembly surface

The objective of this study is to prove that this type of manipulation scheme is possible and contact angle manipulation for a liquid bridge based microgripper is a feasible method of manipulation of objects in microscale.

1.2 Synopsis

The synopsis of the chapters in this dissertation is presented in this section. Chapter II gives a concise literature survey on different microgripper designs along with their advantages and limitations to form the platform of this study. The fundamental aspect of the liquid bridge based microgripper is the shape of the liquid meniscus and the dependence of this shape on different design parameters. In Chapter III the evolution of the shape of the meniscus has been discussed in detail, the important parameters and equations that govern the shape of the liquid bridge have been documented and the
mathematical formulation of the lifting forces and surface energy described from first principle approach. Different approaches for solving the meniscus geometry has been discussed and the numerical algorithm implemented to solve for the meniscus geometry has been presented along with the results and analysis to support the gripping principles. An overview of electrowetting method can be found in Chapter IV. The underlying physics of electrowetting and the limitations of the method are pointed out in detail. The applications of electrowetting in present MEMS technology and how it contributed to this study is demonstrated in this chapter. A commercial software CFD ACE+ is used to solve for the meniscus geometries, and the results are presented in Chapter V. These results are further been used to validate the results obtained in chapter III. Different approaches for the stability study of the liquid bridge are documented in Chapter VI. A summary of the study along with discussions on future research directions can be found in Chapter VII.
2.1 What is a Microsystem?

The present trend in miniaturization of systems has led us to numerous inventions and devices with diverse application. In one hand we have seen the advances in computational power of components made from Integrated Circuit (IC) technology and on the other hand Micro Electro Mechanical Systems (MEMS) technology is booming with devices ranging from accelerometers, oscillators, micromirrors to microfluidic application for biomedical devices. Combination of these technologies eminently shows the prospect of powerful yet complex future inventions which can be termed as Microsystems. It is widely expected that in near future there will be numerous devices that will revolutionize the consumer market. In 1998 NEXUS (The European Network of Excellence in Multifunctional Microsystems) established a task force entitled ‘Market Analysis MST’ to prepare application oriented market analysis from 1996 to 2000. In a survey report [1] the world market of Microsystems was predicted to grown from 14.4 billion Euro in 1996 to 38 billion Euro by year 2002, reflecting a growth rate of 18% per year. In the new report entitled “Market Analysis for MEMS and Microsystems III 2005-
2009” the total market growth for 1st level packaged Microsystems and MEMS is estimated to rise from $12 billion in 2004 to $25 billion in 2009 resulting in an annual compound growth rate of 16%. Incorporating commercially available MST the market will increase from $33.5 billion in 2004 to $57.1 billion in 2009. The top three products available in market are read/write heads, inkjet heads and microdisplays which account for around 70% of the market. Other fast growing markets are microphones, RF MEMS, tire pressure monitoring sensors. Important MEMS devices which are going through extensive research are drug delivery systems, lab on a chip systems, optical switches, magneto optical heads, micro motors, and liquid display. As the devices are becoming complex in nature there is a rise in need to integrate silicon micromachining and CMOS circuits and also incorporating materials like polymer, metals and ceramics in a single device. So the growing need of microsystems is instigating the need of microassembly process for proper integration of micro components.

2.2 What is Microassembly?

Assembly of different microcomponents with submicron level precision is termed as microassembly. As described in [17] “Microassembly lies between conventional macro scale assembly (where the component dimensions are greater than 1mm) and the fast emerging area of nano scale assembly (where the component size is less than 1μm)".
2.3 Why Microassembly is Important?

It is important to understand the utility of microassembly and the limitations in the production process of microparts. MEMS and IC share highly efficient and automated fabrication processes using computer aided design and analysis tools, lithographic pattern generation and micromachining techniques such as film deposition and selective etching. But present MEMS devices use monolithic design where all components are fabricated in a one lengthy sequential process in contrast to various processes used in the manufacturing of IC devices. IC manufacturing technology relies on the use of wide variety of nonstandard process and materials that maybe incompatible to each other. This leads to the conclusion that fabricating a MEMS device and IC chip together is not possible. A possible solution to this problem is microassembly which is the discipline of positioning, orienting and assembling of micron sized components. The goal of microassembly is to provide means to achieve hybrid micro scale devices of high complexity while maintaining high yield and low cost.

2.4 Main Challenges in Microassembly

Automated microassembly poses a list of challenges to the robotic community.

1. Precision – The conventional robots have error margin of few hundred microns, so when dealing with the object size of 100 μm the error would be 100% or more.

So in microscale, submicron level precision is required and the conventional open
loop precision assembly devices fail to perform. Closed loop feedback strategies are required to control the assembly process which means highly sensitive sensors and extremely accurate 3D robotic manipulators need to be built.

2. Vision – It is not possible for the human eye to see in microlevel so real-time visual feedback is needed through microscope or high resolution video cameras for a microassembly platform. As the size reduces, the imaging of the object becomes more difficult and the use of Atomic Force Microscope (AFM), Scanning Tunneling Microscope (STM), or Scanning Electron Microscope (SEM) imaging techniques needs to be considered. Where AFM, STM both are good at scales of 10 to 100 nm, SEM is good for a wide range of 50 nm to 100 μm. A big issue is the trade off between field of view and resolution. Vision may easily get obstructed by the manipulation tools as they are larger than the object. An elegant solution to this problem is the so called ‘eye on the hand’, i.e. a camera is mounted on the arm of a robot.

3. Manipulation tools – Scaling effect creates a difference when any manipulation tool interacts with an object in microscale. Due to scaling effect, forces that are insignificant in the macro world, plays a major part now in the micro world. A conventional pick and place robot relies on the gravity but due to scaling, gravity effect is negligible and thus it faces the problem of adhesion. For example, parts with size less than 100 μm will jump on to the gripper due to adhesion forces and lead to imperfect pickup. Releasing that object from the gripper is also a great challenge. This adhesion forces arises primarily due to capillary forces, surface
tension forces, electrostatic forces and Van der Waal’s forces. While researchers successfully imitated conventional miniature robotic gripper arms, overcoming adhesion forces is still a problem. For parts less than 10 μm in size it might be a better idea to perform manipulation in fluid medium using techniques such as laser trapping and dielectrophoresis.

4. Cost – Most micro parts are produced in batch fabrication process. This massive parallel production mode is the main factor for cost reduction of microdevices. If the assembly of microsystems is performed one by one, be it manually or automated it will increase the production cost. Therefore massive parallel microassembly systems may be required.

2.5 Micro Assembly Systems

Following three types of examples on complete microassembly systems was found from the latest research on microassembly.

Master Slave System – Presently manual assembly uses tweezers and depends on human visual feedback. To automate the same process in micro scale a master slave macro-micro teleoperating systems has been built by Kaneko et al. [2]. The position of the master arm which is manipulated by the operator is scaled down using some transfer function to control the position of a scaled down slave arm, using real time teleoperated visual feedback.
1. **Automatic Assembly machines** – To reduce assembly cost, an automated micro assembly machine was proposed by Zhou et al. [3], which uses a combination of visual and force feedback to control the grasping force. Feddema et al. [4] proposed a CAD–driven technique where the position and orientation is recognized by comparison with synthetic image with realistic effect from CAD.

2. **Assembly by Micro robots** – Faithkow et al. [5] proposed a microassembly with micro robots approach where the micro robots, which are 50-80 mm in size, stand on piezoelectric legs and can move on a glass surface using stick slip principle. With a very fine resolution of 10 nm the robots can move and reach any point of the workplace and use three degrees of freedom grippers for the assembly purpose. The main advantage of this system is the flexibility as each robot can perform different tasks according to their specializations.

2.6 Survey of Adhesion Forces

A typical robotic manipulation scenario is the sequence of operations such as pickup, move and release. In macroscale the gravitational force dominates and robot works against the gravitational force during pickup and uses the gravitational force during placing the object. In microscale (mass less than $10^{-6}$ kg) the gravitational and inertia forces becomes insignificant compared to adhesive forces [7]. These strong adhesive forces poses problem during the pick and release. As the gripper approaches, the object tend to jump off the surface and stick to the gripper due to these adhesive forces. When
the part is to be placed on an assembly surface it may adhere better to the gripper surface than the assembly surface and prevent from accurate release and placement. Typical adhesion forces are electrostatic forces, Van der Waal’s forces and capillary forces. Electrostatic forces arise during charge generation and charge transfer during contact. Van der Waal’s forces appear by instantaneous polarization of atoms and molecules due to quantum mechanical effects. Capillary effects arise from interaction between adsorbed moisture between two surfaces. The balance between these forces depends on the conditions like humidity, temperature, surrounding medium, surface condition, material and relative motion. Tshuchitani [6] have studied the surface forces in microstructures and concluded that the dominant surface forces in micro structure are

1. The liquid bridge force due to capillary condensation of the water when the humidity of the atmosphere around the contacting surfaces is high (over 60% RH)

2. The hydrogen bonding force between water molecules adsorbed on the two surfaces when the humidity is relatively low.

3. The Van der Waal’s force when the adsorbed water molecules on the surface have almost disappeared.

To get an idea of the magnitude of adhesion forces, simple numerical examples of adhesion forces between a spherical object of plane is provided in the next section.
2.6.1 Electrostatic Force

The electrostatic force arises due to charge generation or charge transfer during contact. The approximate electrostatic force between a charged sphere and conducting plane is given by Equation (2.1)

\[ F_{elec} = \frac{(4\pi r^2 \sigma)^2}{4\pi \varepsilon r^2} \]

Where \( \sigma \) is surface charge density, \( \varepsilon \) is permittivity and \( r \) is the radius of the sphere, the distance between the sphere and plane considered to be zero. Figure 2.1 shows a simple sketch of the arrangement of sphere above the plane. The assumed charge density \( \sigma \) is approximately \( 1.6 \times 10^{-6} \) Cm\(^{-2} \) [7].

The force per unit area for parallel plates is given by equation (2.2)

\[ p = \frac{1}{2} \varepsilon |E|^2 = \frac{\sigma^2}{2\varepsilon} \]

At atmospheric pressure and centimeter sized gaps, the breakdown strength of air (30kV/cm) limits the maximum charge density to \( 3.1 \times 10^{-5} \) C/m\(^2 \), or peak pressure of about 50 Pa. In principle conductive grippers can reduce the static charging effects. Presence of native oxide on silicon can also withstand charge up to \( 3.1 \times 10^{-9} \) V/m.
2.6.2 Van Der Waal’s Force

Van der Waal’s force originates due to instantaneous polarization of atoms and molecules when they are set close. It is a much weaker bond than ion and covalent bonds. Neutral atoms and molecules are subject to two distinct forces, an attractive Van der Waal’s force in the long ranges and a repulsive force, the result of overlapping electro orbital referred to as Pauli repulsion in the short range. The simple mathematical model which represents the L-J potential is presented in Equation (2.3)

\[ V(r) = 4\epsilon \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \] 2.3
Where $\varepsilon$ is the well depth and $\sigma$ is the hard sphere diameter in the Angstrom range. The term $\left(\frac{\sigma}{r}\right)^{12}$ describes the repulsive force and the term $\left(\frac{\sigma}{r}\right)^{6}$ describes the attractive part.

Figure 2.2 gives an example of the force diagram for argon dimmer.

![Figure 2.2: Leonard-Jones potential for argon dimer, courtesy Wikipedia.org.](image)

The Van der Waals force for a sphere and plane is given approximately [30] by Equation (2.4)

$$F_{vdw} = \frac{hr}{8\pi z^2}$$

Where $h$ is Lifshitz-Van der Waal’s constant ($10^{-20}$), and $z$ is atomic separation between the surfaces taken as $10^{-10}$ m. When the atomic distance becomes very small, the attractive forces changes into severe repulsive forces. Therefore the minimum distance between surfaces is commonly assumed to be approximately 0.1 nm. The force formulae is assuming atomically smooth surfaces, severe corrections need to be made for rough surfaces. To reduce the effect of Van der Waal’s force $F_{vdw}$, Arai [8] used small pyramids

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of few micrometer width and height at gaps of 10 μm on the gripper surface. The pyramids are made of anisotropic etching of silicon. The second advantage of pyramids was the self discharge possibility due to high electric field strength at the tips.

2.6.3 Surface Tension

The capillary force arises if the adsorbed moisture layer between the sphere and the plane forms a liquid bridge due to close contact. Assuming hydrophilic surface and separation distance much smaller than object radius the maximum capillary force can be approximated by Equation (2.5)

\[ F_{\text{cap}} = 4\pi r \gamma \]

Where \( r \) is the object radius, \( \gamma \) is the surface tension of liquid vapor. The above expression is taken from [32] which is an approximation for maximum capillary force occurring at separation distance zero and for a surface with contact angle zero. Figure 2.3 shows a schematic diagram of liquid bridge formed between two surfaces.

Figure 2.3: Liquid bridge formations at the contact of sphere and a hydrophilic plane surface giving rise to capillary force.
The thin layer of moisture causes a lot of problems including improper functioning of mechanism, unusual release of object from gripper, occurrence of short circuit. The solution consists of removing the water by drying techniques such as critical point drying and freeze drying. Another effective way to avoid sticking is the use of hydrophobic layer. More review of several surface forces can be found in [10].

2.6.4 Gravitational Force

For a spherical part of silicon, the gravitational force $F_{grav}$ would be

$$F_{grav} = \frac{4}{3} \pi r^3 \rho_{si} g$$

Where $\rho_{si} = 2300 \text{ kgm}^{-3}$ is the density of silicon, $r$ is object radius, $g$ is acceleration due to gravity.

2.7 Comparison of Adhesion Forces

Utilizing the above definitions and following the work by Fearing [7] Figure 2.4 was plotted for the comparison of different adhesion forces and gravitational forces as a function of object radius. It can be clearly seen that when the object radius decreases from 1 mm to 1 $\mu$m, capillary force dominates. Van der Waal’s force becomes dominant when the gap between the objects becomes very small (less than 100 nm). Electrostatic force is seen to be the least significant force but it can be argued to be a significant force during manipulation. The gravitational force was found to reduce when the object radius
decreases. Fearing proposed here to perform manipulation in dry or vacuum environment to eliminate the capillary force. Later it was understood that rather than eliminating the adhesion force using it for the purpose of manipulation is a better idea. The only question was how to manipulate the capillary force which will be discussed in the following chapters.

Figure 2.4: Plot of Adhesion forces between a spherical object and a plane as a function of object radius.

2.8 Gripping and Releasing Techniques

In the review paper on microgripping [11] several methods of gripping and releasing have been documented. A few of the mechanisms are given below.
- Releasing the component by positive mechanical engagement [12] [13].
- Injection of a small puff of gas to push the object while removing the gripper [12] [14].
- Mechanical release mechanism with a needle: the needle is used to push the object and as the surface contact area between the object and needle is very small, the gravity becomes dominant again and the object will stay in place when the needle is removed [12] [14].
- Destruction of gripping mechanism. For instance a gripper using surface tension force to pick the object, the object can be released by heating the gripper and evaporating the adhesive liquid [15].
- Freeze tweezers. Using a Peltier device to manipulate the temperature at the probe tip. Freezing the liquid bridge for gripping and liquefying it for release [16].
- Vibration of the gripper to release the object [25].
- Mechanical release by stripping off against a sharp edge [14].
- Using adhesion forces, the adhesion forces between the gripper and object should be greater than the adhesion force between gripper and substrate [14].
- Using a single probe and picking the object using adhesion forces. Releasing the object using a rolling motion of the gripper tip [18].
2.9 Gripping Using Physical Contact

The general design methodology follows the conventional approach of using gripping method using physical contact. Three main types of physical contacts i.e. mechanical grippers, adhesive grippers and vacuum grippers are discussed below. Finally the non contact gripper will be discussed at the end.

2.9.1 Mechanical Grippers

Mechanical grippers are traditionally used to pick and place objects in meso scale. In microscale, large contact forces can damage the object, less forces can loose the object, adhesion problems may cause jumping of object or sticking to the gripper. Keeping in mind these problems many kind of micro grippers are designed. A few will be discussed here.

- Electrostatic gripper: Kim [19] et al. proposed an electrostatically actuated gripper, total length of 400 μm and thickness of 2.5 μm. The gripper was designed to catch various objects including 2.7 μm polystyrene spheres and dried red blood cells. The gripper closes completely by applying a voltage of 45 V. A maximum force of 0.1 μN was recorded at 50 V. Sticking problems were observed and the device was very fragile.

- Hydraulic gripper: Suzumori [20] et al. proposed a hydraulically driven gripper prototype, where one jaw is a flexible pressure chamber which bends when
Pressurized, pressing against the other rigid jaw. The prototype is 18 mm long, 8 mm wide and imparts a maximum gripping force of 2N.

- **Pneumatic gripper:** Peirs et al. [21] demonstrated a prototype which consists of a piston driving both jaws simultaneously via linkages. The prototype has a diameter of 5 mm, is 18 mm long and generates a gripping force of 5N at 10 bars. It has Hall sensor incorporated to measure the gripper opening.

- **Piezoelectric gripper:** Jericho et al. [21] built from patterned polysilicon foil of thickness 2 μm driven by a piezoelectric bar to manipulate bacteria of order ~ 1μm. Haddab [22] proposed another piezoelectric microgripper made up of two piezoelectric bending unimorphs, the thickness of the parts would make it possible to manipulate different sized objects. It has the flexibility to attach various end effectors to manipulate objects with different characteristics.

- **SMA gripper:** Bellouard et al. [23] designed two Smart Material Actuator (SMA) grippers, cut out from a single SMA plate. One prototype uses two ways shape memory effect and the other uses elasticity of structure itself as a bias spring.

- **Thermal gripper:** Greitman et al. used a thermal gripper for one jaw, while the other jaw is fixed but contains piezoresistive force sensors. The gripper is around 1.5 mm long, has a maximum tip displacement of 0.8 mm, generates gripping force up to 1 mN and measures a force resolution of 0.2 μN.
2.9.2 Adhesive Gripper

The sticking forces like Van der Waals Force, Electrostatic Force or the Surface Tension forces can be used to build adhesive grippers. Van der Waals force is relatively small and is difficult to control. Electrostatic force is relatively easy to control but it may not be useful for charge sensitive IC device. Surface tension forces due to high humidity can be controlled using a micro heater at the tip. WestKamper et al. [25] used alcohol as the adhesive liquid instead of using natural moisture for capillary effect. When the gripper is brought in contact with the object the capillary force acts perpendicular to the gripper and surface tension forces offer automatic centering of the component. The gripper demonstrated position tolerance better than $\pm 20 \mu m$ for 4.2 x 4.2 mm$^2$ chips.

2.9.3 Vacuum Gripper

A vacuum gripper is simple as it consists of a thin tube or pipette connected to a vacuum pump. A particular problem is that the tube has to be very thin, and therefore is easily obstructed by small particles. An interesting alternative was to generate vacuum in the gripper itself as shown in [26]. The surface of the tips is covered with micro holes of 10-15 $\mu m$ diameter. The tip is heated before catching and in contact of the object the temperature is decreased resulting in pressure drop inside the micro holes which creates a vacuum to attract the object to the gripper.
2.9.4 Noncontact Manipulation

To overcome the problems associated with the contact between the gripper and object research has been performed on non contact manipulations techniques also. A few of these methods are discussed below.

- **Optical trap or laser tweezers**: The optical pressure that occurs when light is refracted, absorbed or reflected by an object, can be used to manipulate objects of sizes ranging from a few micrometers to few hundred micrometers. To trap an object a focused laser beam with spot diameter of a few microns is aimed at the object. This method can be used in air as well as in liquid medium. A liquid medium has the advantages that the viscous damping increases the traps stability and the buoyancy force helps to levitate the object. For levitating an electrostatic force is best used otherwise the required laser power would cause a heating effect [27]. Morishima et al. [28] successfully used laser tweezers to trap single Escherichia coli bacteria.

- **Magnetic fields**: The objects to be manipulated must be intrinsically magnetic material otherwise small magnetic particles would adhere to the objects. Super paramagnetic particles can be used with high magnetic susceptibility and saturation magnetization but very weak magnetic hysteresis. Such particles act as magnetic dipoles when placed in magnetic field but lose their magnetism when field is turned off. C. H. Ahn et al. [29] used this method to build a micro machined magnetic particle separator.
Electric fields: A particle in an electric field is subject to two types of forces. One is called Coulomb force where two oppositely charged particles are attracted to each other. This effect is also called electrophoresis. The other one is called dielectrophoretic force, which works on an uncharged object and is based on polarization of the object in the electric field. The dielectrophoretic force is proportional to the gradient of the square of the electric field and acts in the direction of the gradient. This allows the transportation of the particles and traps them at spots with high or low field line densities. Electrophoresis and dielectrophoresis are used in biotechnology to separate, trap and classify cells, bacteria, viruses and DNA. Morishima et al. [28] used electric fields to transport E. Coli Bacteria and DNA molecules.
CHAPTER III
THEORETICAL FRAMEWORK FOR SOLVING THE LIQUID BRIDGE MENISCUS GEOMETRY

I shall now proceed to lay before mathematicians another case, which is still more remarkable, from the variety and singularity of the phenomena depending upon it, and from its being susceptible of an equally accurate analysis; the case alluded to is that of capillary attraction.

P.S. Laplace, Tracte de mecanique Celeste (1805)

3.1 Introduction

In the previous chapter from the survey of adhesion forces it was found that the capillary force and the surface tension force are the most important forces in the microscale. The novel idea of using the capillary force as a manipulation tool has inspired researchers to come up with several capillary gripper prototypes and capillary meniscus models. To implement the idea of controlling these adhesive forces first we need to create a framework to obtain the shape of any liquid bridge between two surfaces. So the focus of this chapter is to successfully create a theoretical framework to obtain the meniscus shapes of a liquid bridge between a plate and a sphere. In order to do so we will first
review the important governing parameters and equations used and then define the mathematics needed to solve this problem. Once the shape is obtained the forces will be defined and calculated and a parametric study on the forces would reveal the importance of contact angle manipulation. The study would give us an insight to the feasibility of using liquid bridge for picking up an object and controlling the contact angle for releasing purpose.

3.2 Definition of Parameters

For a systematic approach to solving the problem the important governing parameters need to address in the beginning. In the following paragraphs the concept of curvature of an arc, curvature of a surface, surface tension, contact angle, Young’s equation and Young-Laplace equation is discussed in detail

3.2.1 Curvature

The liquid bridge between two surfaces forms a meniscus profile which is defined by the principle curvatures of the surface. The definition of curvature is thus a very important starting point for the study.
This curvature for a two dimensional curve is defined in [32]. Let $x = x(s)$, $0 \leq s \leq l$ be the function of curve $C$ in terms of arc length $(s)$ as shown in Figure 3.1. The tangent vector $t(s)$ and $t(s + \Delta s)$ at the point $P(s)$ and $P(s + \Delta s)$ with arc length $s$ and $s + \Delta s$ forms an angle $\Delta \alpha$ with one another. If now $\Delta s$ tends to zero, then there exists a limiting value and the curvature of the arc $\chi(s)$ at the point $P(s)$ can be defined as

$$\lim_{\Delta s \to 0} \left| \frac{\Delta \alpha}{\Delta s} \right| = \chi(s)$$

For a straight line one will always have $\Delta \alpha = 0$, that is the curvature $\chi(s)$ is identically zero. The unit of the curvature is therefore the unit of inverse distance. The curvature is therefore a measure of deviation of the form of the curve from straight line.
3.2.2 Principle Curvature of Surface

Any arbitrary curved surface has two principle radii of curvature, meridional curvature and azimuthal curvature as shown in Figure 3.2. The radius of curvature of a surface is complicated but well defined in [33].

![Figure 3.2: Definition of the two principle radii of curvature for any arbitrary surface in x-y plane (a) meridional curvature (1/R₁) (b) azimuthal curvature (1/R₂)](image)

If one erects a normal \( N \) to any surface at a certain point and then passes a plane containing the normal, then the plane intersects the surface and forms a curve as shown in Figure 3.2(a), and the radius of the curve at that point is known as meridional curvature \( (\chi_1) \). The second radius of curvature, known as azimuthal curvature \( (\chi_2) \) can be obtained...
on an orthogonal plane containing the normal as shown in Figure 3.2(b). The intersection of this plane with the surface would give a second curve and a second radius of curvature. If the first plane is rotated through a full circle, the first radius of curvature will go through a maximum and its value at the maximum is called the principle radius of curvature. The second principle radius of curvature is then in a plane orthogonal to the first.

The principle radius of curvature is an extrinsic curvature. The derivation of the form is straightforward from analytical geometry as shown below. If the function of the curve is defined in cartesian coordinates by equations \( x = x(t) \) and \( y = y(t) \) then following the definition of curvature in (3.1) the meridional curvature can be defined as

\[
\kappa_1 = \frac{d\alpha}{ds} = \frac{d\alpha}{dt} \frac{ds}{dt} = \frac{d\alpha}{dt} \frac{1}{\sqrt{x'^2 + y'^2}}
\]

Calculations:

where \( \tan \alpha = \frac{dy}{dx} = \frac{dy/dt}{dx/dt} = \frac{y'}{x'} \)

\[
\frac{d(\tan \alpha)}{dt} = \frac{d(\frac{y'}{x'})}{dt}
\]

\[
\sec^2 \alpha \frac{d\alpha}{dt} = \frac{x'y'' - y'x''}{x'^2}
\]

\[
\frac{d\alpha}{dt} = \frac{1}{(1 + \tan^2 \alpha)} \frac{x'y'' - y'x''}{x'^2}
\]

\[
\frac{d\alpha}{dt} = \frac{x'y'' - y'x''}{x'^2 + y'^2}
\]

\[
\kappa_1 = \frac{x'y'' - y'x''}{\sqrt{x'^2 + y'^2}} = \frac{x'y'' - y'x''}{(x'^2 + y')^{3/2}} \tag{3.2}
\]
where \( x' = \frac{dx}{dt}, \ y' = \frac{dy}{dt} \)

So for a curve defined as \( y = y(x) \) the parametric form can be written as \( x = t, \ y = y(t) \)

\( x' = 1, \ x'' = 0 \) and substituting it to Equation (3.2) gives the expression for the curvature in cartesian coordinates as (3.3)

\[
\frac{1}{R_1} = \kappa_1 = \frac{d^2 y}{dx^2} \left[ 1 + \left( \frac{dy}{dx} \right)^2 \right]^{\frac{3}{2}}
\]

The second principle radius of curvature is in a plane perpendicular to the plane of the principle radius and generally obtained by extending the normal to the profile until it touches the axis of revolution. The expression for \( R_2 \) can be obtained by trigonometric relation i.e. \( \sin \alpha = \frac{x}{R_2} \)

\[
\frac{1}{R_2} = \kappa_2 = \sin \alpha = \tan \alpha = \left( \frac{dy}{dx} \right) x \sec \alpha \left[ 1 + \left( \frac{dy}{dx} \right)^2 \right]^{\frac{1}{2}}
\]

For a sphere both these curvature are positive and equal to each other.

3.2.3 Sign Convention of the Radius of Curvature

In the definition of mean curvature given by Finn [34] he pointed out that the curvature should be considered positive when the curve is bending in the direction of
normal vector $N$. In another paper by Orr [35] it is cited that the curvature should be considered positive when its center of curvature lies in the axis side of the meniscus

### 3.2.4 Mean Curvature

For any arbitrary curved surface the mean curvature is defined as the arithmetic mean of the two radii of principle curvatures $\chi_1$ and $\chi_2$ as shown in Equation (3.5)

$$H = \frac{1}{2}(\chi_1 + \chi_2) = \frac{1}{2}\left(\frac{1}{R_1} + \frac{1}{R_2}\right)$$  \hspace{1cm} 3.5

### 3.2.5 Surface Tension

Surface tension is defined in two distinct ways. One way we can see surface tension as a force per unit length of a surface and on the other hand it can be considered as surface energy per unit area. The term surface tension implies that an actual state of tension exists between the molecules of the material on the surface. Molecules inside the material phase stays in force equilibrium state as the surrounding molecules exert attraction force on them. Molecules on the surface get exposed to the outside environment which disrupts the natural force equilibrium resulting in extra state of tension from the surrounding molecules from inside the material phase. This extra force per unit length is called surface tension. On the other hand if we consider energy per unit area concept then surface tension is the energy required to create unit area of surface by bringing molecules from the interior of the phase to the surface region. Surface tension is
responsible for the shape of the liquid droplet. The cohesive forces on the surface layer
tend to form a shape with minimum surface energy. The magnitude of surface tension
depends on the physical properties of the media in contact.

The following two cases may arise when liquid having a free surface comes in
contact with a rigid body:

1. Complete wetting, when the entire surface of the rigid body is covered either by
   the liquid or by its thin film.

2. Partial wetting, when the liquid covers only a part of the surface of a rigid body.
   Here the liquid forms a certain angle at the wetting line, which is called the
   contact angle or the wetting angle.

3.2.6 Contact Angle

Contact angle is the quantitative measurement of the wetting of a solid by liquid.
It is the angle formed by the liquid at the three phase boundary of solid, liquid and gas. It
is detected by a tangent drawn at the triple point, the point at which the three phases
intersect as shown in Figure 3.3.
Ideally the contact angle is expressed by the Young’s Equation as in (3.6)
\[
\cos \theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}}
\]  
(3.6)

where $\gamma_{lv}$, $\gamma_{sv}$, $\gamma_{sl}$ are the surface tensions at the liquid – vapor, solid- vapor, and solid - liquid boundaries and $\theta$ is the contact angle. The shape of drop and magnitude of contact angle is controlled by the interfacial tensions of each participating phases. In non ideal conditions surface roughness, electric field or chemical heterogeneity leads to deviations from this relationship. It is often necessary to distinguish between the static contact angle as defined above and the dynamic contact angles. i.e. advancing contact angles and receding contact angles. The presence of dynamic contact angle accounts for contact angle hysteresis.
3.2.7 Laplace Equation

The Laplace equation is the fundamental equation of capillarity. To characterize the rise of liquid in capillary tube quantitatively Laplace showed that the mean curvature of the free surface is proportional to the pressure change across the surface. Though Fin [34] termed his reasoning too simplified and not entirely convincing, it has now become the standard presentation in engineering textbooks. For a small section of arbitrary curved surface with two radii of curvature \( R_1 \) and \( R_2 \), Laplace equation is expressed as (3.7)

\[
\Delta p = \gamma_h \left( \frac{1}{R_1} + \frac{1}{R_2} \right)
\]

where \( \gamma_h \) is the surface tension of the liquid, \( \Delta p \) is the pressure difference across the interface. The pressure difference is defined as (3.8)

\[
\Delta p = p_i - p_o
\]

where \( p_i \) is the pressure inside the liquid phase or inner pressure and \( p_o \) is the pressure on the side of the gas phase or outside pressure. Positive pressure difference means the pressure inside the liquid is higher than the pressure outside which accounts for a convex meniscus shape. Negative pressure difference means the pressure inside the liquid is lower than the outside pressure and meniscus shape is concave in nature.
3.2.8 Derivation of Laplace Equation

A simple proof for Laplace Equation from the concept of work done was presented in the book written by Adamson [33]. A small section of arbitrary curved surface is taken as shown in Figure 3.4. The two radii of curvature $R_1$ and $R_2$ are indicated in the figure and the section taken is small enough so that $R_1$ and $R_2$ are essentially constant. If the surface is displaced by a small distance outwards, ignoring the smallest terms $dxdy$ the change in area will be

$$\Delta A = (x + dx)(y + dy) - xy \approx xdy + ydx$$  \hspace{1cm} 3.9

Figure 3.4: Derivation of Laplace equations for arbitrary surfaces

The work done in forming this amount of surface is then given by Equation (3.10)

$$Work = \gamma_n (xdy + ydx)$$  \hspace{1cm} 3.10
Due to a pressure difference $\Delta p$ across the surface the work done to move the area $xy$ through a distance $dz$ against the pressure is (3.11)

$$\text{Work} = \Delta p xy dz$$  

3.11

By comparing similar triangles it follows that

$$\frac{x + dx}{R_1 + dz} = \frac{x}{R_1} \quad \text{or} \quad dx = \frac{x dz}{R_1}$$

$$\frac{y + dy}{R_2 + dy} = \frac{y}{R_2} \quad \text{or} \quad dy = \frac{y dz}{R_2}$$  

3.12

If the surface is in mechanical equilibrium, the two work terms must be equal so equating them and substituting the expression for $dx$ and $dy$ gives an equation of the form (3.13)

$$\Delta p = \gamma \left( \frac{1}{R_1} + \frac{1}{R_2} \right)$$  

3.13

Thus the Laplace equation can be derived from the work done concept.

### 3.2.9 Kelvin Equation

For a drop of liquid in equilibrium with vapor it is useful and common to derive another equation which is usually called Kelvin equation. The Kelvin equation gives the relation between a spherical liquid vapor surface with mean radius of curvature $R$ in equilibrium with the atmosphere and is defined as (3.14)

$$\Delta p = \frac{RT}{V_m} \ln \frac{P_0}{P_s}$$  

3.14
where \( v_m \) is the molecular volume of the liquid, \( p_0 \) is the pressure outside the drop, \( p_s \) is the saturated vapor pressure of the atmosphere, \( R \) is the ideal gas constant (8.3 x 10^7 erg/deg.mol). Kelvin equation can be derived in a number of ways and several approximations are introduced in various ways but the most elegant way to derive it is given by Rowlinson et al. [38]. From the thermodynamic point of view the chemical potential is considered to be same for both inside and outside of the drop whether it has finite radius \( R \) or infinite radius which leads to the derivation of Kelvin equation (3.15) with three assumptions

\[
kT \ln \frac{p_0}{p_s} \approx \frac{1}{\rho_i} \left( \frac{2\gamma}{R} + p_m - p_s \right)
\]

3.15

The assumptions are

i) The liquid is incompressible

ii) The vapor is a perfect gas

iii) \( \frac{2\gamma}{R} \gg p_0 - p_s \)

where \( p_m \) is the pressure in the liquid side, \( p_s \) is the saturated vapor pressure of vapor and the definition of \( p_0 \), the pressure outside the drop is ambiguous which has been discussed by Powles [39]. Powles further stated that Kelvin’s equation is valid for temperatures approaching critical temperature and for microscopic drop as far as homogeneous thermodynamics is valid. It should be noted that Kelvin equation is important for problems where the liquid bridge is formed due to condensation of vapor from the atmosphere when two surfaces are brought in close proximity to each other.
3.3 Modeling the Meniscus Profile

The meniscus profile can be obtained by solving for the Young-Laplace equation. The Young-Laplace equation relates to difference in hydrostatic pressure across the interface to the local mean curvature $H$ (notation $H$ will be used instead of $\chi$ in this section) and surface tension $\gamma$ (used instead of $\gamma_b$). Mathematically this is a nonlinear two point boundary value problem and different approaches have been used to solve for this problem. The three prominent methods used are presented in this chapter

1. Solving in terms of elliptic integrals, Orr et al.[35]
2. Solving the two point boundary value problem by shooting and bisection method
3. Solving through Circular or parabolic approximation methods

These different solutions are discussed in the following sections.

3.3.1 Analytical Solution of Young-Laplace Equation Using Elliptic Integrals

Orr et al. [35] solved the Young-Laplace equation analytically in terms of elliptic integrals for all possible types of pendular rings between sphere and plate considering the effect of gravity to be negligible. The types of meniscus have been categorized by their shape, i.e. meniscus profile having zero mean curvature is catenoid, other shapes like nodoid, unduloid, spheroids and their properties have also been discussed. The expressions for the forces associated with the capillary and surface tension have been formulated and studied extensively. The occurrence of capillary repulsion where it
successfully overcomes the surface tension force was also analyzed. In this paper for the first time the general case of a sphere in contact with a flat plate, and the meniscus making different contact angles with the plate and the sphere has been considered. Finally they proposed a circular approximation for the general sphere and plate problem and studied its range of validity.

Before getting into the solution, the Bond Number should be defined as it plays a major role in the following discussions. The Bond Number is defined as the ratio of the gravitational force with the surface tension force (3.16)

$$B = \frac{\rho g L^2}{\gamma}$$

where $\rho$ is the density of the liquid, $g$ is the gravitational acceleration, $L$ is a characteristic length taken as the radius of a spherical droplet or the length of a cylinder depending on the type of problem and $\gamma$ is the surface tension force of the liquid in vapor. Sometimes the density is replaced by $\Delta \rho$, which is the density difference between fluids on the either side of the meniscus. When $B<<1$, the effect of gravity is negligible. In the following section the method of solution described by Orr is discussed.

Figure 3.5 shows the meniscus profile of a liquid bridge between a plate and a spherical object of radius $R$ along with all the important parameters. The liquid bridge is considered axisymmetric with contact angles $\theta_1$ with the flat surface, $\theta_2$ with the spherical surface, the objects are separated by a distance $D$.

If the meniscus is axisymmetric in $rz$ coordinate system then the mean curvature satisfies the following Young-Laplace Equation (3.17)
\[
\frac{\Delta p}{\gamma} = 2H = \frac{1}{R_1} + \frac{1}{R_2} = \frac{d^2z}{dr^2} + \frac{dz}{dr} \left[ 1 + \left( \frac{dz}{dr} \right)^2 \right]^{\frac{3}{2}} \left[ 1 + \left( \frac{dz}{dr} \right)^2 \right]^{\frac{1}{2}} \tag{3.17}
\]

With dimensionless variables \( y = \frac{z}{R} \) and \( x = \frac{r}{R} \) and the parameter \( u = - \sin \varepsilon \), where \( \varepsilon \) is the angle made by the normal to the meniscus with vertical axis the Laplace equation reduces to the form (3.18)

\[
2HR = -\frac{du}{dx} - \frac{u}{x} \tag{3.18}
\]

Figure 3.5: Meniscus profile of liquid bridge formed between a sphere and a plane
Equation 3.18 must be solved as a two point boundary value problem for a known $\Delta p$ to find out the meniscus profile. The boundary conditions are the contact angles at the contact line of liquid and solid surfaces stated as in (3.19)

$$u_1 = -\sin(\pi - \theta), \quad y_1 = 0 \quad x_1 = \sin \phi$$

$$u_2 = -\sin(\theta + \phi), \quad y_2 = d + 1 - \cos \phi,$$

Where $\phi$ is the half filling angle and $d = D/R$ is the normalized separation between sphere and plate as shown in Figure 3.5.

The boundary value problem has the solution [35]

$$x = \left(\frac{1}{2H}\right) \left[ u \mu \left( u^2 + c \right)^{\frac{1}{2}} \right]$$

$$y = \frac{1}{2HR} \int_{u_1}^{u_2} \left( \frac{u du}{1 - u^2} \right) \left[ \frac{u \mu \left( u^2 + c \right)^{\frac{1}{2}}}{\mu \left( u^2 + c \right)^{\frac{1}{2}}} \right]$$

Where the curvature dependent parameter $c$ is

$$c = 4H^2 R^2 \sin^2 \phi - 4HR \sin \phi \sin(\theta_1 + \phi)$$

The mean curvature $H$ needs to be solved through an iterative solution of equation (3.23)

$$2HR = \left(\frac{1}{\mu} \right) \int_{y_1}^{y_2} \left( \frac{u du}{1 - u^2} \right) \left[ \frac{u \mu \left( u^2 + c \right)^{\frac{1}{2}}}{\mu \left( u^2 + c \right)^{\frac{1}{2}}} \right]$$

The complicated aspect of this equation is that the signs need to be chosen, and the choice is dictated by the sign of the meridional curvature of the meniscus. In (3.18) the meridional curvature is $-du/dx$ and the azimuthal curvature is $-u/x$. From (3.20) the meridional curvature can be expressed as
An elaborate guideline on the process of solution has been documented in this paper. A very important contribution of this paper is a table containing all the formulae associated with the curvatures, surface areas and enclosed volume.

Later this process of solution was found in a paper by Obata et al. [36], where they proposed a micromanipulation method based on volume control of a liquid bridge. They pointed out that Orr has solved the Young-Laplace equation analytically in terms of elliptic integral, but did not give a solution for the problem where the volume of the liquid bridge can be kept constant. Their work furthered the solution to problem with volume constraint and the assumptions for their solutions were as follows:

i) The influence of gravity is negligible i.e. the Bond Number $B \ll 1$

ii) The dynamic flow of the liquid is negligible

iii) The volume of the liquid is conserved i.e. no evaporation and condensation of liquid is occurring

iv) The contact angles were determined by Young’s equation

v) The object and the plate are rigid

vi) The area of the plate is infinite

A table was provided as a guide to choose the sign for the equations. Normalized volume $v \ (V / R^3)$ and the normalized distance $d \ (D / R)$ were also defined in terms of integral of $\varepsilon$
\[ v = \int_{z_1}^{z_2} \pi x^2 \, dz = \int_{\epsilon_1}^{\epsilon_2} \pi x^2 \frac{dz}{d\epsilon} \, d\epsilon \]  

3.25

and

\[ d = z_1 - (1 - \cos \phi) \]  

3.26

The attractive force due to meniscus was defined as the sum of the pressure difference and the axial component of the surface tension acting on the object

\[ F = 2\pi R \gamma \left[ \sin \phi \sin(\theta_i + \phi) - HR \sin^2 \phi \right] \]  

3.27

The manipulation scheme used there was control of forces by changing the liquid volume for the liquid bridge. The contact angle was kept the same for both the liquid sphere and liquid plate contact line.

This process was found to be tedious and complicated and other process of solutions were further studied for solving the Young-Laplace equation.

3.3.2 Solving Young-Laplace Equation Using Iterative Method

It was found from Aveyard et al. [37] that the nonlinear problem can be expressed in a parametric form and solved by a shooting and bisection method.
Figure 3.6: Meniscus profile of liquid bridge formed between a flat gripper surface and the spherical object.

For our case in order to obtain the meniscus geometry, the nonlinear differential equation should be solved with contact angles as the boundary conditions and constant volume as a constraint. The volume of the liquid should be kept constant at the desired value. The gap $D$ is considered to be known and constant. The liquid bridge is considered to be axisymmetric and gravitational effect and viscous effect is neglected in the simulation.
We used a triple iterative scheme to solve for the geometry. Dimensionless variables were employed where the radius of the sphere was used as a characteristic length \( L (L = R) \) as shown in (3.28)

\[
R = \frac{R}{L} \quad D = \frac{D}{L} \quad r = \frac{r}{L} \quad z = \frac{z}{L} \quad V = \frac{V}{L^3}
\]

The pressure difference is scaled as \( \Delta \zeta = \Delta p \, L \). The scaled pressure difference is used in solving the nondimensional parametric Laplace equation. It should be noted that \( \Delta p \) is used for actual pressure difference and later this parameter is used as a control parameter for iteration scheme, but \( \Delta \zeta \) is the scaled pressure difference, and the it is used in the solving the parametric Laplace equation. Laplace Equation in parametric form is expressed by Equation (3.29)

\[
\frac{dr}{ds} = \cos \alpha \; ; \; \frac{dz}{ds} = \sin \alpha \; ; \; \frac{d\alpha}{ds} = \frac{\Delta \zeta \gamma - \sin \alpha}{r}
\]

where \( s \) is the length of meniscus arc in the capillary bridge geometry and \( \alpha \) is the angle of the normal with the \( z \) axis at any point on the meniscus as shown in Figure 3.6. To find the characteristics of a bridge for given \( r_i \) and \( \Delta p \) values the differential equation (3.29) is numerically integrated by a fourth order Runge-Kutta method with initial conditions (3.30)

\[
\alpha(0) = \pi - \theta_i \; , \; \quad r(0) = r_i \; , \; \quad z(0) = 0
\]
3.3.3 Numerical Algorithm for Solution of Laplace Equation at Constant Volume

The iterative scheme is described in this section. A schematic diagram of the flowchart is presented in Figure 3.7 and a sample picture of the iteration process in Matlab is presented in Figure 3.8. The values of the parameters needed at the start of the iteration are object radius $R$, contact angles $\theta_1$, $\theta_2$, the constrained volume of the liquid $V_c$ and a known gap $D$.

1. For any initial value of contact radius $r_1$ and pressure difference $\Delta p$ the equation (3.29) can be solved for any arc length $s$ satisfying the initial conditions in (3.30).

![Figure 3.7: Flowchart for the triple iterative algorithm to solve for the parametric Young-Laplace equation (3.29)](image)

48
So a shooting method was used and the values of arc length $s$ was increased from zero in steps of $\Delta s$, up to the point where the arc touches the surface of the sphere. At each value of $s$ the solution of 3.29 gives out the values of $r_2$, $z_2$ and $\alpha_2$. The arc touches the sphere when $z_2 = H_2$ where $H_2$ is defined as in 3.31

$$H_2 = D + R_2 - \sqrt{R_2^2 - r_2^2}$$

Initially $z_2 < H_2$, but when $z_2 > H_2$, a relative error function defined as $\frac{H_2 - z_2}{H_2}$ changes its sign. Then a bisection method was used to locate the touching point $(r_2, z_2)$ where $z_2 = H_2$, with a precision of $\varepsilon_s$. Once the end point of the curve is found the half filling angle $\psi$ can be calculated by simple trigonometric equation $\psi = a \sin \left( \frac{r_2}{R_2} \right)$ and contact angle $\theta_f$ can be calculated by the
equation $\theta_f = \alpha_2 - \psi$. If it fails to touch the surface and diverges away from the surface the shooting and iteration method on $s$ need to be repeated with a different initial value of $r_1$ or $\Delta p$.

2. The next step is to satisfy the boundary condition of contact angle $\theta_2$. An angle error function was defined as $\theta_f - \theta_2$ and a shooting method was implemented on the parameter $\Delta p$. Here we have used a control parameter $P_r$ defined as $P_r = \frac{P_0}{P_s}$ and obtained the values of $\Delta p$ through Kelvin equation (3.14). The parameter $P_r$ was incremented in steps of $\Delta P_r$ to a point where the error function changes sign. A bisection method was used to minimize the error function to some permissible error $\varepsilon_\theta$. As a result the final value of $P_r$ and also $\Delta p$, which satisfies the boundary condition of $\theta_f = \theta_2$ can be obtained. If the boundary condition cannot be satisfied the initial value of $r_1$ or $P_r$ needs to be changed.

3. Finally we need to satisfy the volume constraint. The volume of the axisymmetric meniscus profile is calculated using the equation (3.32)

$$V_f = \int_0^{z_2} \pi r^2 dz - \frac{\pi}{3} R^3 (2 + \cos\psi)(1 - \cos\psi)^2$$

A volume error function was defined as $\frac{V - V_f}{V}$ and a shooting method was implemented on the contact radius $r_1$. The contact radius $r_1$ is incremented by a value $\Delta r_1$ to a point where the volume error function changes sign. Then a
bisection method was used to minimize the error to some permissible error value of $\varepsilon_V$. As a result the final value of parameters $r_1$ and $P_r$ can be obtained.

It was understood that the selection of the initial values and the increment values are very important for obtaining a solution through this iteration process. It was also understood that the solutions obtained were not unique and for different sets of initial values, the solutions differs. To tackle this problem an approach was taken where the parameters $r_1$ and $P_r$ were incremented and decremented simultaneously from their initial values to find the multiple positions where the error function changes sign. For a number of problems two solutions were obtained through this method and both the solutions were preserved for further analysis. The process of three nested iteration of shooting and bisection method is found out to very time consuming and a proper choice of increment values should be found out by $\Delta r_1$ and $\Delta P_r$ by trial and error method for each volume operation which can commensurate the time but without compromising the accuracy of the solutions.

3.4 Characterization of Energies

The energy characterized by Finn [34] is the most comprehensive so far. He argued that Laplace Equation and Young Equation, though accepted by the scientific community as a standard is not completely flawless. Both these equations are derived through the minimization of Total Energy as shown in a number of publications. The
definition of the energy terms is crucial to understand these equations completely. So in his book he characterized energy from the viewpoint of Gauss, where Gauss based his reasoning on the principle of virtual work, according to which energy of a mechanical system in equilibrium is unvaried under arbitrary virtual displacements with the constraints. For general three phase system consisting of fluid, gas and rigid walls he conveniently divided the energy in question into three terms

1. **Free Surface Energy** – If the configuration is to be in equilibrium, the elements of a fluid in the free surface separating two media must be more attracted to that fluid than to outer medium (fluid or gas), otherwise the two media would mix and the surface would disappear. Thus there is a differential attraction and resulting lowering of fluid density within the surface. The energy associated with this removal of fluid from the surface must be proportional to the surface area which can be written as

\[ E_s = \gamma S \]  

Where \( \gamma \) the surface tension and \( S \) is the surface area. For the different phase interaction and the corresponding common areas the free surface energies can be individually calculated.

2. **Wetting Energy** – This is the adhesion energy between fluid and the rigid wall. It is analogous to the surface energy, except that the fluid particles near the rigid surface can experience the larger attraction in either direction (since the walls are rigid, the surface cannot disappear when the net attraction is towards the wall). The energy can be written as
\[ E_w = -\gamma \beta S^* \] \hspace{1cm} 3.34

Where \( S^* \) is chosen to be any portion of the wetted surface that includes the neighborhood of the contact line. The sign on the right of Equation 3.34 is chosen such that \( \beta > 0 \) corresponds to a wetting configuration. Finn showed that the condition \( |\beta| \leq 1 \) is necessary for stability of the configuration. \( \beta \) has been referred as relative adhesion coefficient between fluid and the bounding walls on \( S^* \).

3. Gravitational Energy – For general purpose the potential energy is considered \( Y \) per unit mass and the resultant energy is an integral taken over all of the space that is occupied by the media.

\[ E_y = \int Y \rho \, dx \] \hspace{1cm} 3.35

\( \rho \) is the difference between the local fluid density and the density of the outer medium. The domain of the integration can be restricted to any region in which the variations have its support, the density exterior to the fluid being taken to be zero.

The total energy is the summation of all these energies terms. We have not considered the energy associated with the gravitation here. The definition of interfacial energy was obtained from [42]. It states that "When two immiscible liquid 1 and 2 are in contact the free energy change in expanding their interfacial area by unit area is known as their interfacial energy or interfacial tension \( \gamma_{12} \). The energies associated with this expansion process may be understood by splitting it into two hypothetical steps; first the
areas of media 1 and 2 are created and then brought into contact”. The total free energy change $\gamma_{12}$ is therefore

$$\gamma_{12} = \gamma_1 + \gamma_2 - W_{12}$$

where $W_{12}$ is the energy required to separate the two areas in media 1 and 2 in. Equation (3.36) is often referred to as Dupree’s equation. Following the same rule we can calculate the total energy associated with a liquid bridge between two surfaces by three hypothetical steps as shown in Figure 3.9.

![Figure 3.9: Three hypothetical steps for calculation of Surface Energy (a) free surfaces of the liquid bridge (b) liquid surface attached to flat surface $A_1$ (c) liquid surface attached to both the surface, flat surface $A_1$ and curved surface $A_3$. The mediums 1, 2, 3, and 4 correspond to solid1, liquid, solid 2 and vapor/gas.](image)

From step 1, the energy required to create the liquid surfaces of the liquid bridge is found to be $\gamma_{24}(A_1 + A_2 + A_3)$. Now some amount of work is needed to go from Figure
3.9(b) to 3.9(a), i.e. to separate the liquid from the solid flat surface, that amount of work must be subtracted from the energy term and the energy equation for step 2 is
\[ \gamma_{24} (A_1 + A_2 + A_3) - W_{142} A_1 \]
following the same rule for Figure 3.9(c), we get the final expression for the energy as
\[
\gamma_{24} (A_1 + A_2 + A_3) - W_{142} A_1 - W_{243} A_2
\]
where \( W_{142} \) is the energy required to separate medium 1 and 2 in medium 4 expressed as
\[ W_{142} = \gamma_{14} + \gamma_{42} - \gamma_{21} \] and \( W_{243} \) is the energy required to separate medium 2 and 3 in medium 4 expressed as
\[ W_{243} = \gamma_{24} + \gamma_{43} - \gamma_{32} \] .
Expressing the mediums 1, 2, 3, and 4 as solid1, liquid, solid2 and vapor/gas and substituting the values to the total energy equation, substituting young’s equation and after simplification the energy term was expressed as (3.38)
\[
E = -A_1 \gamma_{lv} \cos \theta_1 - A_3 \gamma_{lv} \cos \theta_2 + A_2 \gamma_{lv}
\]
where \( A_1 \) is the surface area between liquid and sphere interface, \( A_2 \) is the surface area of the liquid vapor interface, \( A_3 \) is the surface area of liquid and plane interface, \( \gamma_{lv} \) is the surface tension of the liquid in vapor.
3.5 Formulation of Force

The total force exerted through the bridge on the sphere and the plate consists of three parts, a surface tension force which originated from the line of contact of liquid and the solids, a capillary force which is transmitted by the liquid but originates in the curvature of the meniscus and is due to the pressure difference across the meniscus, and a buoyancy force associated with the wetted segment of the sphere and the plate. The third force is neglected as the bond number is small so effectively we have only two forces. There are two sets of forces. If we consider the liquid bridge and the object together then the forces acting on the gripper surface will have two components. The axial component of the surface tension force $F_{s1}$ (negative if attractive) acting on the liquid is given by (3.39)

$$F_{s1} = -(2\pi r_1) \gamma_v l_v \sin(\theta_1)$$

Figure 3.10: Free body diagram of the Lifting forces acting on the gripper surface and the surface of the spherical object.
where $2\pi r_1$ is the circumference of the contact line of the liquid at the planar surface and
$\gamma_b\sin(\theta_1)$ is the axial component of the surface tension. The axial component of the capillary force $F_{c1}$ is defined in (3.40)

$$F_{c1} = \Delta p \pi r_1^2 = 2C\gamma_b \pi r_1^2$$

where $\pi r_1^2$ is the effective area on which the capillary pressure acts.

If we consider the forces acting only on the object at the liquid sphere interface the axial component of the surface tension force $F_{s2}$ is given by (3.41)

$$F_{s2} = -(2\pi R \sin \psi) (\gamma_b \sin (\theta_2 + \psi))$$

where the circumference of the contact line of the liquid and sphere interface is $2\pi R \sin \psi$ and $\gamma_b \sin (\theta_1 + \psi)$ is the axial component of the surface tension.

The axial component of capillary force $F_{c2}$ is defined in (3.42)

$$F_{c2} = \Delta p \pi (R \sin \psi)^2 = 2C\gamma_b \pi R^2 \sin^2 \psi$$

where $\pi (R \sin \psi)^2$ is the effective area on which the capillary pressure acts and $\Delta p$ is the pressure difference across the interface. If $\Delta p$ is negative the pressure inside the liquid bridge is less than the pressure outside which creates a low pressure inside which effectively produces an attractive force on the sphere.

Hence the total force $F_{t1}$ acting on the sphere and liquid bridge at the planar gripper surface is (3.43)

$$F_{t1} = F_{s1} + F_{c1} = -2\pi \gamma_b r_1 [\sin(\theta_1) - Cr_1]$$

The total force $F_{t2}$ on the sphere exerted by the liquid bridge is summed up in (3.44)

$$F_{t2} = F_{s2} + F_{c2} = -2\pi \gamma R [\sin \psi \sin(\theta_2 + \psi) - CR \sin^2(\psi)]$$
If we consider the free body diagram of the liquid bridge these two forces $F_{t1}$ and $F_{t2}$ as shown in Figure 3.10 should be acting in opposite directions and for an equilibrium state when gravitational effect is neglected they should be equal. Numerical results obtained later have proved this point and all the lifting forces analyzed later is $F_{t1}$.

Depending on the meniscus geometry the value of $\Delta p$ can be positive, zero or negative. When $\Delta p$ is zero the capillary force becomes zero and only the surface tension force remains. As the pressure difference changes sign the two forces act in opposite direction and at a certain point the total lifting force vanishes. Beyond that there is repulsive force acting on the sphere.

3.6 Validation of Numerical Scheme

In order to validate the accuracy of the numerical solution, the results were matched against an analytical solution. The Young-Laplace equation can be solved analytically for zero pressure difference resulting in a catenary meniscus profile [35]. The analytical equation for such a profile is expressed as in (3.45)

$$r = A \cosh\left(\frac{z - B}{A}\right)$$

Where for $0 \leq \theta_1 \leq \pi/2$, $A = a \sin (\theta_1)$, $B = -a \cosh (a/A) A$ and for $\pi/2 \leq \theta_1 \leq \pi$, $A = -a \sin (\theta_1)$, $B = -a \cosh (a/A) A$ and $a$ is the value of the contact radius $r_1$. The iteration solution matches accurately with a catenary curve as shown in Figure 3.11 for two different contact angles 60° and 120°.
Figure 3.11: Comparison of the meniscus geometry obtained from the iterative solution of Laplace equation for $\Delta p = 0$ and $r_f = 0.1$ and catenary curve ($\square$). The parameters used here are dimensionless $R = 1$, $D = 0.05$, $\theta_2 = 20^\circ$ and (a) $\theta_1 = 60^\circ$ (b) $\theta_2 = 120^\circ$.

3.7 Validation of Lifting Force

Experimental study on measurement of capillary attraction force for a liquid bridge based microgripper was performed and published by Lambert et al [8]. The data from this paper has been used to validate our algorithm. Their experimental setup measured the attraction force of a silicon oil liquid bridge between a flat surface and spherical surface of radius 7.9 mm ($\theta_1 = 16^\circ$, $\theta_2 = 12^\circ$). The surface tension of silicon oil is 0.0208 N/m and the volume is 0.5 $\mu$l. The simulation results matches well for larger gap but for small gap below 0.2 mm our simulation gave larger forces.
3.8 Simulation Results

Numerical simulation was performed by changing the gaps between a spherical object and a flat surface for different volumes. We will consider a spherical object as a sphere of glass bead with contact angle 20°. For each study in this section the radius of the object is considered to be 1 mm. As the parameters were non-dimensionalized by the radius of the object $R$, the dimensionless radius in the results is always $R = 1$, the dimensionless volumes used are 1.0, 0.1, 0.01 and 0.001. For convenience it is noted that for an object of radius 1 mm the volumes corresponds to 1.0 μl, 0.1 μl, 0.01μl, and 0.001μl. The contact angles are taken as $\theta_1 = 60^\circ$ and $\theta_2 = 20^\circ$. The solution of Laplace equation (3.30)
using the iteration method as described Section 3.3.3 gives one and more than one solution depending the parameters used.

3.8.1 Meniscus Profile for Change in Gap

The effect of gap on the meniscus profile shape is studied in this section. It was found that for smaller values of gap there is one solution and when the gap is increased there are two solutions. As the gap reaches a critical value the solutions converges to each other and after it reaches the critical value no further solution was achieved which satisfies the boundary conditions. Figure 3.12 shows an example of meniscus profile geometries for volume of 0.1, when the gap is changed from 0.1 to 0.48.

![Figure 3.12](image)

Figure 3.12: Meniscus profile geometries for different dimensionless gaps (a) $D = 0.1$ (b) $D = 0.2$ for dimensionless liquid volume of 0.1, contact angles $\theta_1 = 60^\circ$ and $\theta_2 = 20^\circ$ and object radius of 1 mm.
Figure 3.13: Meniscus geometries of two solutions of the Laplace equation (—— stable, 
-·-·-· unstable) for different dimensionless gaps (c) $D = 0.3$ (d) $D = 0.4$ (e) $D$
= 0.45 (f) $D = 0.48$ for dimensionless liquid volume of 0.1, contact angles $\theta_1$
= 60° and $\theta_2$ = 20° and object radius of 1 mm.(continued).

It was found in paper by Lian et al. [40] and also in Erle. et al. [41] that Young-
Laplace equation has two solutions and as we also noticed in Figure 3.12, the two
solutions converged to a single point beyond which no solutions exist. This point was
defined as the rupture point of the liquid bridge. Following their definition of stable and
unstable bridges, the free surface energies associated with these two solutions were
calculated and the solution with lower surface energy is termed as a stable solution and
that corresponding to higher surface energy as an unstable solution. Figure 3.13 shows the surface energy profiles obtained for dimensionless volumes of 0.1, 0.01 and 0.001. The rupture points have been pointed out. Figure 3.14 show that the lifting forces are decreasing for increase in gap. An interesting observation was noted for dimensionless volume of 1, for lowest gap 0.002, the force is less than the force obtained from volume 0.1 and the slope of the decrease in force is flatter than other volumes.

Figure 3.14: Plot of Surface Energy of the liquid bridge for increasing gap for volumes 0.1, 0.01 and 0.001, contact angle $\theta_1 = 60^\circ$ and $\theta_2 = 20^\circ$ and object radius of 1 mm. (—— stable, – – – unstable)
Figure 3.15: Plot of Total Lifting Force $F_t$ of the liquid bridge for increasing gap for volumes $0.1, 0.01$ and $0.001$, contact angle $\theta_1 = 60^\circ$ and $\theta_2 = 20^\circ$ and object radius of $1$ mm. (— stable, – – – unstable).

3.8.2 Meniscus Profile for Change in Contact Angle

A set of simulations were performed for different volumes to investigate the effect of contact angle changes on the meniscus profile, surface energy and lifting forces. The first step in manipulation is the pickup phase. When the gripper with a liquid droplet on its surface approaches the object, the liquid surface comes in contact with the object and forms a liquid bridge. The object attaches to the gripper and the gap between the spherical object and the gripper surface would be crucial starting point of the simulations. Since the gravitational effect is neglected the liquid droplet height ($d$) of Figure 3.15 is taken as the gap between the object and the gripper surface ($D$) for further simulations.
Figure 3.16: Height (d) of the pendant droplet on the gripper surface

The liquid droplet height was calculated from the simulated droplet geometry in CFD ACE+ software for contact angle $\theta_1 = 60^\circ$, and was found to be 0.042 mm, 0.09 mm, 0.2 mm, and 0.46 mm for volumes 0.001 $\mu l$, 0.01 $\mu l$, 0.1 $\mu l$, and 1 $\mu l$, respectively. We have a constraint in the angle range so the contact angles used here is greater than $60^\circ$. The contact angle is increased with an increment of $10^\circ$ to the critical point after which no solution can be obtained. An increment of $1^\circ$ has been used in the area of the critical region. The other contact angle is kept constant at $\theta_2 = 20^\circ$. The focus of the study was to find out if the liquid bridge can be ruptured by increasing the contact angle at the time of release of the object. Figure 3.13 shows the different meniscus profiles obtained when the contact angle is increased. The liquid bridge volume used here is 0.1 $\mu l$. It was noted that for higher contact angles two solutions were obtained and the solutions converge this angle can be considered a critical angle corresponding to rupture. Figure 3.17 shows the surface energy profiles and Figure 3.18 shows the lifting forces for different volumes. when the angle is further increased. Figure 3.16 shows an example of evolution meniscus profiles for dimensionless volume 0.1. Following the previous discussion for gap change.
Figure 3.17: Meniscus geometries for two solutions for different contact angle $\theta_1$ c)

100°, (d) 120° (e) 140° (f) 144° for dimensionless volume of 0.1 with contact angle $\theta_2 = 20^\circ$ and object radius of 1 mm. (—— stable , –·–·– unstable (continued),
Figure 3.18: Plot of Evolution of the surface energy with change in contact angle $\theta_1$ for dimensionless volumes of 0.1, 0.01 and 0.001, contact angle $\theta_2 = 20^\circ$. (—— stable, – – – unstable).

Figure 3.19: Plot of total lifting force with the change in contact angle $\theta_1$ for different dimensionless volumes of 0.1, 0.01 and 0.001, contact angle $\theta_2 = 20^\circ$. (—— stable, – – – unstable).
From the Figure 3.16 it was found out that by changing the contact angle the lifting forces can be reduced below the weight of the object. For volume 0.001 the lifting forces were not significant with respect to the weight of the object. The forces are significant for volumes of 0.01, 0.1 and 1. All of the volumes cross a critical angle of rupture in the region of 145° to 150°. If our device have a hydrophobic surface of 120°, and by electrowetting the angle can be reduced to 70° only then from this plot a volume range between 0.001 to 1.0 can chosen to be the appropriate working volume. A dense plot of forces for different volumes can be used for choice of volume for our manipulation scheme.

Figure 3.20: Plot of total lifting force with the change in contact angle $\theta_1$ for different dimensionless volumes of 0.01, 0.1 and 1.0, contact angle $\theta_2 = 20^\circ$ and gap $D = 0$, and for dimensionless volume 0.1 , gap $D = 0.2$. 

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For any liquid volume when the liquid bridge is formed between the two surfaces the equilibrium gap that could be seen cannot be predicted with any analytical equation. A dynamic study can provide some insight into the stable equilibrium gap of the liquid bridge. The assumption of gap taken in this study can face many challenges and it was found that the assumption of zero gap could be a more acceptable assumption. Simulation was performed for zero gap i.e., D=0, and the contact angle was changed from 60° to 120°. It has been found that the lifting force still decreases below the weight of the object for the dimensionless volumes 0.01, 0.1 and 1.0. It is noted that the slope of the decrease of force for zero gap is higher than that the slope decrease of force for gap 0.2 (dimensionless). This study shows that for zero gap the lifting force is much greater and also by change in contact angle the lifting force decreases fast, facilitating a quick release of the object.

3.9 Circular Approximation of Meniscus Shape

To avoid the complications of the solution procedure of Laplace equation by elliptic integral method or iteration method, an approximation model can be used to substitute the meniscus geometry. Previous works [35] [43] have demonstrated that the meridional curvature can be approximated by the radius of a circle and therefore the axisymmetric liquid bridge is approximated by a toroidal approximation. Figure 3.21 shows the geometry of the circular meniscus between the spherical object and the lat surface.
Figure 3.21: The circular approximation diagram for a liquid bridge between a spherical surface of radius \( R \) and a flat surface at a distance \( D \), where liquid have a contact angle \( \theta_1 \) with the flat surface, and \( \theta_2 \) with the spherical surface and the liquid is wetting the spherical surface with a half filling angle \( \psi \).

In this context the Laplace equation in equation (3.4) can be written as

\[
\Delta p = \gamma \left( -\frac{1}{r_1} + \frac{1}{r_2} \right) \quad 3.46
\]

where \( r_1 \) and \( r_2 \) are the meridional and the azimuthal curvature respectively as shown in Figure 3.21. The sign convention of the values of \( r_1 \) and \( r_2 \) should be kept in mind when the mean curvature is calculated. In general when a concave meniscus is considered the
center of curvatures lies on either side of the curve and the meridional curvature is negative and azimuthal curvature is positive. For a convex profile both the centers of curvature lies on the same side of the curve and both can be taken as positive values. From the equation (3.46) it can also be observed when \( r_2 \gg r_1 \) the mean curvature can be approximated by only the meridional curvature as shown in (3.47). For small values of gap \( D \) and large volume of the liquid this approximation is valid.

\[
\Delta p = -\frac{\gamma}{r_1}
\]

It should also be noted here the Laplace equation corresponds to a constant mean curvature, and if the meridional curvature is considered to be a constant radius of a circle, the azimuthal curvature should also be defined as a constant term. But by definition the azimuthal curvature can take different values at different points on the circular curve. Three different definitions of azimuthal curvature were found in [43]

\[
r_2 = \frac{R \sin \psi}{\sin(\theta_2 - \psi)} \quad \text{Maximum possible value of } r_2
\]

\[
r_2 = R \sin \psi \quad \text{Intermediate value of } r_2 \quad 3.48
\]

\[
r_2 = x_2 - r_1 (1 - \sin(\theta_2 - \psi)) \quad \text{Minimum possible values of } r_2, \text{ radius of the neck}
\]

The last definition is known as Gorge method and from [40] it was found that this definition produces errors in Force calculation within 10% of the force calculated by numerical techniques. In our study we have used the intermediate value. The Laplace equation or the azimuthal curvature does not govern the shape of the meniscus here and the meniscus geometry can be easily found by simple trigonometric
manipulations. So for a set of known parameters i.e., radius of the spherical object \( R \), gap between the two surfaces \( D \), contact angles \( \theta_1 \) and \( \theta_2 \), and half filling angle \( \psi \), the unknown values of contact point of liquid at the spherical surface \((x_2, y_2)\), meridional curvature \( r_1 \), azimuthal curvature \( r_2 \), center point of the circular meniscus \((x_c, y_c)\) and contact radius on the flat surface \((x_1, y_1)\) can be obtained by the equations (3.49)

\[
\begin{align*}
x_2 &= R \sin \psi \\
y_2 &= R + D - R \cos \psi \\
r_1 &= \frac{R + D - R \cos \psi}{\sin \left( \frac{\pi}{2} - \theta_2 - \psi \right) + \sin \left( \frac{\pi}{2} - \theta_1 \right)} \\
r_2 &= R \sin \psi \\
x_c &= x_2 + r_1 \cos \left( \frac{\pi}{2} - \theta_2 - \psi \right) \\
y_c &= r_1 \cos \theta_1 \\
x_1 &= x_c - r_1 \sin \theta_1 \\
y_1 &= 0
\end{align*}
\]

To find the meniscus geometry with a volume \( V_c \), a shooting and bisection method was implemented on the half filling \( \psi \) and the solution was found where \( V = V_c \).

The volume of the liquid bridge was calculated by the equation (3.50)

\[
\begin{align*}
V &= \pi_c (x_c - r_1)^2 y_2 + 2 \pi_c r_1 y_c - \frac{\pi}{3} \left( (y_2 - y_c)^3 + y_c^3 \right) - \pi_c r_1^2 (\pi - \theta_1 - \theta_2 - \psi) \\
&\quad + \pi_c (y_2 - y_c)(x_2 - x_c) - \pi_c y_c (x_c - x_1) - \frac{\pi}{3} R^3 (2 + \cos \psi) (1 - \cos \psi)^2
\end{align*}
\]

The solution was then compared with the solutions obtained from the numerical iteration method.
Figure 3.22: Comparison of stable solution from numerical solution and circular approximation of meniscus shape of a liquid bridge of dimensionless volume 0.01, between a flat surface and a spherical surface with dimensionless radius $R = 1$ with a gap $D = 0.09$, contact angles $\theta_2 = 20^\circ$ and different values of $\theta_1$

(a) 60° (b) 80° (c) 100° (d) 120° (e) 140°.
Figure 3.23: Comparison of stable solution from numerical solution and circular approximation of meniscus shape of a liquid bridge of dimensionless volume 0.1, between a flat surface and a spherical surface with dimensionless radius $R = 1$ with a gap $D = 0.20$, contact angles $\theta_2 = 20^\circ$ and different values of $\theta_1$ (a) $60^\circ$ (b) $80^\circ$ (c) $100^\circ$ (d) $120^\circ$. 
Figure 3.24: Comparison of stable solution from numerical solution and circular approximation of meniscus shape of a liquid bridge of dimensionless volume 1.0, between a flat surface and a spherical surface with dimensionless radius $R = 1$ with a gap $D = 0.46$, contact angles $\theta_2 = 20^\circ$ and different values of $\theta_1$ (a) $60^\circ$ (b) $80^\circ$ (c) $100^\circ$.

From the Figures 3.22 – 3.24 it can be observed that the circular approximation matches the shape obtained from the numerical solutions very efficiently for smaller volumes and smaller contact angles. As the contact angles increases discrepancy in shape
arises as the meridional curvature becomes very large and for further increase in angle, the solver is unable to provide any solution.

3.10 Conclusion

This Chapter aims at understanding the surface tension forces and capillary forces and forming a mathematical framework to obtain the meniscus geometry for further investigations. A short overview of different methods shows the complications of the computation method involved for solving this problem. The Laplace equation was finally solved with boundary conditions and volume constraint by a triple iterative algorithm. The algorithm was validated against an analytical solution and they matched perfectly. The results for change in gap and change in contact angle were obtained. An interesting observation has been the non uniqueness of the solution of Laplace equation. The convergence of two solutions and the critical values were obtained for different volumes of operation. Finally it can be concluded that the simulation results supports the hypothesis that contact angle manipulation can be implemented for liquid bridge based microgripper application. There are some design challenges which need to be considered in future work. The initial configuration of the droplet and the object when the liquid bridge carries the object needs further attention and an energy based study would provide further information on this area. The mathematical modeling of circular approximation of the meniscus geometry is discussed and it was shown that for smaller angles (60°-100°)
and smaller dimensionless volume (0.01 and 0.1) the circular approximation is matching the solution obtained by rigorous iteration scheme very well.
CHAPTER IV

ELECTROWETTING

4.1 Introduction

In recent years Electrowetting has become an effective method in comparison to others as it has been proven to be very successful in manipulating liquid droplet on a surface and liquid in micro channels. It was understood that electrowetting can be used to control the capillary and surface tension forces by manipulation of contact angle. This chapter focuses on understanding the underlying theory of electrowetting, its limitations like saturation of contact angle reduction, the evolution of application areas of electrowetting in micro technologies and last but not the least, design of an electrowetting mechanism for our device.

4.2 Electrowetting

The electric control of liquid surfaces demands special attention among different phenomena investigated in electro hydrodynamics. Electrowetting is essentially a phenomenon where an electric field can modify the wetting behavior of a liquid in
contact with a solid electrode. As a definition Electrowetting is the change in solid liquid contact angle due to an applied potential difference between the solid and the liquid.

4.2.1 History of Electrowetting

Electrocapillarity, the basis of modern electrowetting was first described in detail in 1875 by Gabriel Lippman [44] (see translation of the original Lippman’s paper [44] in Mugele and Baret (2005) [46]). He first observed that capillary depression of mercury in contact with electrolytic solutions could be varied by applying a voltage between mercury and electrolyte. His theory states that “the capillary constant at the mercury / diluted sulphuric acid interface is a function of electrical difference at the surface” which explains his understanding that electrowetting originates from the change in the solid-liquid interface energy. The capillary constant above refers to the interfacial tension term. The major problem in broader application of electrocapillarity was the electrolytic decomposition of water upon application of voltages beyond few hundred millivolts. While these fundamental phenomena were discovered a long time ago it was only approximately two decades ago, that researcher and engineers realized the vast practical potentials of these findings.
4.2.2 Electrowetting On Dielectric (EWOD)

The recent developments were initiated by Berge [45] in early 1990’s when he introduced the idea of using a thin insulating layer to separate the conductive liquid from metallic electrode in order to eliminate the problem of electrolysis.

![Water droplet on hydrophobic surface](image)

Figure 4.1: Water droplet on hydrophobic surface (a) without voltage and (b) with voltage, courtesy Philips [55]

This effect was called Electrowetting on Insular Coated Electrodes (EICE) [45]. Later this configuration is often called electrowetting-on-dielectric (EWOD). In the Figure 4.1 an example of electrowetting is displayed, where voltage is applied between the electrode in the water and electrode beneath the hydrophobic surface. The application of voltage has changed the contact angle of the water.
Initially, electrowetting applications included crude oil purification and ink jet printing but recently the vast prospect of this technology has been understood in the light of micro technologies and it is being considered as the primary mechanism for manipulating small amounts of fluid. A number of applications in micro-scale was cited by (Mugele and Baret 2005[46], Darhuber and Trojan 2005[56]), applications include lab-on-the-chip (Pollack et al. 2002[57]), adjustable lenses (Berge and Peseux 2000[58]) and electronic displays [55].

4.2.3 Young Lippman’s Equation

Young’s equation relates the interfacial energies of three phase in an interphase with the contact angle and is expressed by the equation 4.1

\[ \gamma_v \cos \theta = \gamma_{sv} - \gamma_{sl} \]  \hspace{1cm} 4.1

where \( \theta \) is local contact angle of the liquid with the solid, \( \gamma \) is the interfacial energies for the different phases and the subscripts \( s, l, v \) stands for solid, liquid and vapor. Young’s equation can be derived from a balance of all forces acting on the contact line as shown in Chapter 3. When electrical potential is applied across the interface of solid and liquid, the voltage applied generates electric charges on surfaces on the solid and liquid. The mechanism of charge generation, and its distribution, can be rather complicated for real electrolytes, but the assumption of an ideally conducting fluid with a surface charge density is usually sufficiently accurate. The presence of this surface charge modifies the contact angle, which can be estimated from the principle of minimum energy (Berge
or by combining the Young’s and Lippman’s equation leading to the Young-Lippman’s formula 4.2

\[ \cos \theta = \cos \theta_0 + \frac{\varepsilon_0 \varepsilon_1}{2d \gamma_{lv}} U^2 \]  

where \( \varepsilon_0 \) is permittivity of vacuum, \( \varepsilon_1 \) is the permittivity of the liquid, \( d \) is the thickness of the ionic double layer or the thickness of the dielectric layer, \( \theta_0 \) is the contact angle in normal conditions without application of electric potential and \( \theta \) is the contact angle after applied potential of \( U \). The potential for zero charge is considered zero.

An identical equation can be derived using the thermodynamic or electromechanical approach (Mugele and Baret 2005[46]) discussed later. Considering all simplifying assumptions, (4.4) agrees with the experimental data surprisingly well for a range of contact angles. Many authors validated this theoretical prediction for different droplet fluids and electrode sizes, dielectric film materials and thickness, voltage levels, as recorded in (Berge 1993[48]; Vallet et al. 1999[54]; Verheijen and Prins 1999[49]; Decamps and De Coninck 2000[59]; Moon et al. 2002[60]).

4.3 Theoretical Background

In this section the basic aspects of electrowetting will be discussed first and then short descriptions of the different theoretical approaches to tackle this problem will be presented.
Electrowetting deals with droplets partially wetting a surface and changes in contact angle with change in applied voltage. As shown in Figure 4.2 the droplet resides on an electrode with a dielectric coating of thickness $d$. In Lippman’s experiment the electrical potential was applied between the liquid and the electrode. In most applications the droplet size is of the order of 1mm radius or less. For liquids of this order the Bond number is smaller than unity. The Bond Number $B_0 = \sqrt{\frac{g \rho r^2}{\gamma_{lv}}}$ is a measure of strength of gravity with respect to the surface tension, $\rho$ is the difference in density of the liquid, $r$ is the radius of the droplet and $\gamma_{lv}$ is the surface tension of liquid in vapor. All along this study the effect of gravity has been neglected. The ambient condition can be considered as air or vapor.

Figure 4.2: Generic Electrowetting on Dielectric (EWOD) Setup
Electrowetting has been studied by researchers from various fields such as applied physics, chemistry and engineering discipline. Depending on the backgrounds different approaches were used to describe the electrowetting phenomenon. Mugele and Baret [46] presented discussions on three approaches in their review. The three main approaches are:

1. Classical thermodynamics approach
2. Energy Minimization approach
3. Electromechanical approach

4.3.1 Classical Thermodynamics Approach

The classical thermodynamics approach is proposed by Lippman and is based on general Gibb’s interfacial thermodynamics [47]. Lippman’s initial experiment dealt with direct metal electrode and electrolyte interface. Upon application of voltage difference \( dU \), an electric double layer builds up spontaneously at the solid liquid interface consisting of charges on the metal surface on one side and a cloud of charged counter ions on the liquid side of the interface. This interaction reduces the interfacial tension \( \gamma_{sl} \) and the effective interfacial tension becomes dependent on the change in voltage difference \( (dU) \) and charge density \( \sigma \) of the counter ions

\[
d\gamma_{sl}^{\text{eff}} = -\sigma dU
\]

For simplicity it is assumed that the counter ions are all located at a distance \( d_H \) (of the order of few nanometers) from the surface (Helmholtz model). The double layer has a
fixed capacitance \( c_H = \varepsilon_0 \varepsilon_1 / d_H \), where \( \varepsilon_1 \) is the dielectric constant of the liquid. The effective interfacial tension is now expressed as below

\[
\gamma_{sl}^\text{eff} (U) = \gamma_{sl} - \int_{U_{pzc}}^U \rho_{sl} dU = \gamma_{sl} - \int_{U_{pzc}}^U c_H UdU = \gamma_{sl} - \frac{\varepsilon_0 \varepsilon_1}{2d_H} \left( U^2 - U_{pzc}^2 \right)
\]

4.4

Here \( U_{pzc} \) is the potential at zero charge. Metal electrodes when immersed in electrolyte produces spontaneous charging and the potential required to compensate that charge is \( U_{pzc} \). Using Young’s equation along with the equation the change in contact angle is described as

\[
\cos(\theta) = \cos(\theta_0) + \frac{\varepsilon_0 \varepsilon_1}{2d_H \gamma_{lv}} \left( U^2 - U_{pzc}^2 \right)
\]

4.5

This equation is applicable for a voltage range below the onset of electrolytic process.

Modern application of electrowetting usually circumvents this problem by introducing a thin layer of dielectric material between the metallic electrode and the liquid. The electric double layer is developed at the interface of the dielectric and the liquid. As an advantage the liquid need not be an ionic solution. The insulator acts as a capacitor \( (c_d = \varepsilon_0 \varepsilon_1 / d) \) in series with the capacitor formed through the ionic double layer. As the insulator thickness \( d \) is much larger that the double layer thickness \( d_H \), \( c_d \ll c_H \), neglecting \( c_d \) the equivalent series capacitance can be written as

\[
c_{eq} = \frac{1}{c_d} + \frac{1}{c_H} = \frac{c_d c_H}{c_d + c_H} \approx \frac{c_d c_H}{c_H} \approx c_d
\]

4.6

The modified equation becomes
\[ \gamma_{sl}^{\text{eff}}(U) = \gamma_{sl} - \frac{\varepsilon_0 \varepsilon_1}{2d_H} U^2 \]  

4.7

Here it is assumed that the surface of the insulating layer does not give rise to spontaneous adsorption of charge in the absence of an applied field, i.e. \( U_{pzc} = 0 \).

Combining with Young’s equation it is known as Young- Lippman’s equation

\[ \cos(\theta) = \cos(\theta_0) + \frac{\varepsilon_0 \varepsilon_1}{2d_H \gamma_{lv}} U^2 = \cos(\theta_0) + \frac{\eta}{2} U^2 \]  

4.8

where \( \eta \) is a new dimensionless electrowetting number which measures the strength of electrostatic energy compared to surface tension. The thickness of the dielectric is larger than the thickness of the double layer as a result the capacitance of the dielectric is less than the capacitance of the double layer. As the total capacitance of the system is approximated as the capacitance of the dielectric medium it reduces tremendously for this modified version and the voltage required to decrease the contact angle is much higher in EWOD.

From the Equation 4.9 the order of magnitude analysis would give us

\[ U \approx \sqrt{\frac{\gamma_{lv}}{c}} = \sqrt{\frac{d_H \gamma_{lv}}{\varepsilon_0 \varepsilon_1}} \]  

4.9

Where capacitance \( c \) being \(~100 \text{ pF/cm}^2\) for EICE, while in conventional studies with bare metal \( c \) is at least \( 10^4 \) times larger [45]. Thus instead of 100 mV needed for bare electrode the driving voltage becomes a few hundred volts, which is allowable as it remains less than the breakdown threshold of the dielectric material. As the working range would be far below the breakdown voltage the dielectric will undergo little degradation due to applied voltage which is a great benefit for practical purpose.
change in contact angle is around a few tens of degrees to 80 degrees. Recent experiments with different dielectric media have reduce the voltage requirement to as low as 15-30 Volts.

4.3.2 Energy Minimization Approach

The energy minimization approach for electrowetting was first formulated by Berge [48]. The total energy of a droplet in an EWOD system is the summation of the interfacial energy term $F_{if}$ and the electrostatic energy term $F_{el}$. The electrostatic contribution is defined as

$$F_{el} = \frac{1}{2} \int \vec{P} \cdot \vec{D} dV$$ \hspace{1cm} 4.10

Where $\vec{E}(\vec{r})$ and $\vec{D}(\vec{r}) = \varepsilon_0 \varepsilon(\vec{r}) \vec{E}(\vec{r})$ denotes the electric field and electric displacement at $\vec{r}$, $\varepsilon(\vec{r})$ is the dielectric constant of the medium at location $\vec{r}$. The corresponding total energy can be written as

$$F = F_{if} - F_{el} = \Sigma A_i \gamma_i - \Delta p V - \frac{1}{2} \int \vec{P} \cdot \vec{D} dV$$ \hspace{1cm} 4.11

For a droplet the electrostatic energy can be divided into two parts. The first part arises due to the presence of a capacitor between the electrode and the liquid and the second part is due to capacitance along the edge of the droplet line of contact. The second part is known as fringe field effect and it is neglected. So the total electrostatic energy can be written as
\[ F_{el} = \frac{CU^2}{2} = \varepsilon_0 \varepsilon_d U^2 A_{sl} / 2d \]

The negative sign in the energy can be understood intuitively by considering the entire system comprising of the droplet and the power supply (battery). In that case the energy gain due to electrowetting is taking place in the battery and excess charge is being redistributed from the battery to the electrode surface. For a homogeneous electrode the free energy now can be expressed as

\[ F = A_{lv} \sigma_{lv} + A_{sv} \sigma_{sv} + A_{sl} \left( \sigma_{sl} - \frac{\varepsilon_0 \varepsilon_1 U^2}{2d} \right) - \Delta pV \]

Comparing the coefficient and combining the modified solid liquid surface energy with Young’s equation would produce the Young-Lippman’s equation as we obtained in the last approach.

The assumptions in this derivation is

(i) the interfacial energies are voltage independent

(ii) the liquid is perfectly conductive

(iii) Accumulation of excess charge at the edges or edge effect can be neglected

Verheijen, Prins [49] discussed about the fringe field effect and gave some analytical expressions for stray capacitance for a droplet on a substrate.

4.3.3 Electromechanical Approach

The methods discussed above do not give a physical picture of contact angle reduction. Jones [50] first proposed electromechanical approach taking into consideration
the forces exerted on the liquid by the electric field. For isotropic liquids the electrical forces are formulated in terms of the Kortweg-Helmholtz body force density as shown in Equation (4.14)

\[
\frac{\rho}{f_k} = \rho_j E - \frac{1}{2} E^2 \nabla \varepsilon + \nabla \left[ \frac{1}{2} E^2 \frac{\partial \varepsilon}{\partial \rho} \rho \right]
\]

4.14

where \(E\) is the electric field, \(\rho_j\) is the volume density of free electric charge, \(\rho\) is the mass density and \(\varepsilon\) is the permittivity of the liquid. Electrical force density formulations are not unique in classical continuum electrodynamics but Equation (4.14) can be selected for problems with homogeneous and incompressible liquid. For homogeneous and incompressible liquid the third term that is electrostriction, has no influence on hydrostatics and can be neglected [50]. The net force acting on a volume element \(dV\) can be obtained by volume integration over the previous equation. As a fundamental consequence of momentum conservation the same force can be obtained by integration along the surface of \(dV\) over the momentum flux density of the electric fields, i.e. Maxwell Stress Tensor. Neglecting electrostriction [51] the Maxwell Stress Tensor consistent with Equation 4.14 can be written as Equation 4.15

\[
T_{ik} = \varepsilon_0 \varepsilon_1 \left( E_i E_k - \frac{1}{2} \delta_{ik} E^2 \right)
\]

4.15

Where \(\delta_{ik}\) is Kronecker delta function and \(i, k = x, y, z\). The net force acting on the liquid the liquid volume is a surface integral of the stress tensor in the outward direction. The Maxwell stress is maximal at the contact line and decays to a practically negligible value away from the Triple Contact Line (TCL). The field and charge distribution is found by
solving Laplace equation for an electrostatic potential $\phi$ with appropriate boundary conditions.

Integrating the horizontal component of Maxwell stress, we obtain net force acting on the droplet as

$$F_x = \frac{\varepsilon_0 \varepsilon_1}{2d} U^2 = \sigma n \eta$$  \hspace{1cm} (4.16)

This expression can be used in the force balance at the contact line in the spirit of Young as a result we discover the same Young-Lippman’s equation again. The ideas and derivations can be found in Jones et al. [50] [51]. The shape independence of the force also implies that the contact angle reduction and the force should be regarded as independent phenomenon. As pointed out by Jones [52] the observed change in apparent contact angle and the net electrostatic force producing displacement is two different observations and the change in contact angle does not provide the force for displacement of the liquid.

### 4.4 Contact Angle Saturation

From Young-Lippman’s equation it was thought that by application of potential the contact angle can be reduced to zero, but surprisingly all researchers found that after increasing the potential the contact angle gradually saturates and fails to reach to zero degree. The degree of saturation and the value of saturated contact angle vary widely and depend on the materials used and the experimental setup used. The actual reason for this saturation has puzzled many researchers and a suitable clarification has not been found.
yet. In the following paragraphs some of the saturation models are described which would provide us with some idea of the causes of saturation.

4.4.1 Vallet’s Theory

The experimental and theoretical investigations by Vallet et al. (1999) [54] intended to look into the limiting voltage of the electrowetting phenomenon for a droplet placed on an electrode coated with an insulator film. When using pure water they observed surface instability and emission of small droplets from the contact line. For higher voltage the smooth movement of the contact line stopped and suddenly micro droplets started emitting from the mother droplet. They roughly observed that the measured wavelength of the first unstable mode is directly proportional to the thickness of the insulator film.

Figure 4.3: Experiments on electrowetting and electrowetting saturation, (a) formation and expulsion of micro droplet at the onset of saturation, Mugele et al. [46] (b) & (c) luminescence at the contact edges due to ionization of air at the onset of saturation, Vallet et al. [54].
Their observation was later reproduced Mugele and Herminghaus [46] for mixtures of water and glycerol as shown in Figure 4.3 (a). In another set of experiments with salt solutions, the angle change is observed to stop suddenly without any droplet ejection, but a luminous ring was noticed at points close to the contact line. The concomitant observation of luminescence, apparent spikes in current and contact angle saturation lead them to suggest that the saturation is occurring due to ionization of air around the contact line of the droplet. Later they derived the capacitance of the model using the conformal mapping of the edges which resulted in a modified Young’s equation in the light of edge correction to the electrostatic energy. The modification terms showed that the contact angle change is dependent on the Line tension effect. Furthermore they attempted to explain the saturation under the linear stability theory and formed the limit of stability graphs which was at par with their experimental results.

4.4.2 Verheijen and Prins Theory

Verheijen and Prins 1999[49] proposed two electrical methods for measuring the contact angle and the wetting velocity. The contact angle measurement was based on the capacitance measurement and the radial velocity of the spreading droplet was calculated by measuring the current flowing through the droplet. A very important observation was that the electrowetting is not dependent on the polarity of the applied voltage. Later Verheijen and Prins [76] developed a theory in order to explain the saturation phenomenon in term of charge trapping in or on the dielectric layer. Their assumption
was that a homogeneous layer of charge with constant surface charge density is trapped inside the dielectric material at a certain distance from the electrode which is lowering the electric field at the solid-liquid interface. With these assumptions they derived a modified version of the Young-Lippmann’s equation from the concept of virtual displacement and the modified equation is expressed by equation below

\[ \cos \theta = \cos \theta_0 + \frac{\varepsilon_0 \varepsilon_1}{2d \gamma_{lv}} (U - U_T)^2 \]  

4.17

Where \( d \) is the thickness of the dielectric layer, \( \varepsilon_0 \) is the permittivity in vacuum, \( \varepsilon_0 \) is the permittivity of the dielectric material , \( U_T \) is the potential of the trapped charged layer outside the droplet i.e \( \gamma_T = \frac{\varepsilon_0 \varepsilon_1 U_T}{d_1} \), where \( d_1 \) is the distance of the charged layer from the electrode surface. To explain this concept from the microscopic origin they proposed that the ions on the liquid-solid interface travels through the nano-pores of the dielectric medium due to electrostatic attraction force of the opposite charges ions on the electrode surface. As no dependence of the saturation on the type of ions, valence and polarity of the potential is observed experimentally, it was concluded that this theory needs further research and the microscopic origins cannot be explained conclusively.

4.4.3 Kang’s Theory

Kang (2003) [70] also investigated the electrowetting phenomenon from the viewpoint of classical electrostatics and placed special attention to the influence of excess
charge at the edge region. He followed the conformal mapping technique used by Vallet [54] and analyzed the electrostatic field around the wedge shaped liquid contact region.

![Image](image.png)

**Figure 6.** Electrostatic force and its influence on horizontal balance of forces acting on the three-phase contact line.

**Figure 3.** Charge distribution around the wedge for \( \theta = 60^\circ \).

![Image](image2.png)

**Figure 4.4:** Classical Electrostatic analysis by Kang [70] revealed (a) the electrostatic force acting on the contact line and (b) the charge distribution at the contact edges.

From the results of their study the surface charge density at the contact point was found to be very high relative to that at the solid liquid interface region. Moreover the electrostatic net force was found to be always directed outward, perpendicular to the liquid surface at the contact line, providing a horizontal and vertical component of forces. It was suggested that the horizontal force contribution resulted in Lippman’s modification of the Young’s equation, but the unaccounted vertical force might have an important role to play in the saturation as for small angles this force sharply increases and might resist further decrease of contact angle resulting in saturation. Finally it was concluded that the
electrowetting phenomenon is originating from the electrostatic forces rather than the change in interfacial tension of the solid liquid interface.

### 4.4.4 Shapiro Theory

Shapiro et al. (2003)[63] proposed a total energy minimization based approach with volume constraint to quantify different physical effects on electrowetting. Their analysis is similar to Digilov[77] but they considered a wide range of physical parameters and rigorously transformed all the energy terms into modification terms in Young’s equation. They considered bulk energy like gravitational potential energy and interfacial potential energy and electrical energies like potential energy in solid dielectric layer, potential energy stored in external charging source, energy due to ionic double layer. In order to explain the saturation phenomenon, the authors included in their mathematical model the electrical resistance of the fluid and despite enormous differences between resistivity of the fluid and dielectric, the results seem to confirm their hypothesis.

### 4.4.5 Papathanasiou Theory

The results in Papathanasiou and Boudouvis (2005)[53] seem to indicate that the contact angle saturation can be attributed to the dielectric breakdown of the dielectric solid on which the liquid is placed. Although in Janocha et al. (2000)[61] this possibility
was ruled out. The motivation for the above theory was provided in Seyrat and Hayes (2001) [65], who linked the saturation effect to a material deficiency.

4.4.6 Summary of Saturation Theories

In summary, it is a widespread belief that different physical effects can cause the saturation phenomenon, depending on the configuration of the investigated set up. While some authors claim that electrowetting is responsible for microactuation of droplets, and that some minimum contact angle change is needed for successful microfluidic operations, Moon et al. (2002)[60] and Jones (2002)[67] proved that the net force on the liquid can be exerted only in a non-uniform electric field and is basically dielectrophoretic. For a liquid between two parallel electrodes, the net force is not related to the shape of the meniscus, which depends on the liquid–solid contact angle. A very similar analysis was performed by Kang et al. (2003)[68], who concluded, that ‘wetting phenomenon is primarily a consequence of stress acting on the droplet surface’. However, this can be more complicated in the EWOD droplet actuation applications: the net dielectrophoretic force acting on the droplet depends on the distance between the droplet and the driving electrode (with an electric potential different from the droplet potential). Due to the electrowetting effect the droplet is first flattened, which reduces the distance, leading to an increase of the force. The entire process can be explained without resorting to the deformation of the droplet (Washizu 1998) and theoretically should work for nondeformable spheres as well.
Another point of contention is related to the interpretation of the contact angle change. While some authors take it literally as the local contact angle at the triple, solid–liquid–gas, contact line, it is more and more accepted that this angle should be taken on a macroscopic, or mesoscopic (Mugele and Baret 2005[46]), scale, meaning sufficiently far from the contact line (Shapiro et al. 2003[63]). The earliest proof of that was given in (Buehrle et al. 2003[69]), where the shape of the liquid surface and the electric field near the contact line were calculated at the same time. Although a 2D Cartesian model was assumed, it was shown that the local contact angle at the triple point approaches Young’s value, even for high voltages, when the apparent contact angle far from the contact line is small.

Another interesting conclusion from this work is that there is no well-defined distance from the contact line where the apparent contact angle is reached. Kang (2002) [70] analyzed the electric field distribution near the contact line analytically using the conformal mapping technique. Correctly attributing the changes of the apparent contact angle to electric forces, he calculated the electric field distribution and the force exerted on the entire contact area, not just the contact line, showing exact agreement with the Lippman–Young predictions. A different approach was presented in Buehrle et al. (2003) [69]. For a perfectly conducting wedge defined by an angle $\alpha$, the electric field behaves as $1/r^{-\nu}$, where $\nu = (\pi - \theta)/(2\pi - \theta)$ so it is singular when $r \to 0$. Therefore, the electric pressure diverges as well. However, the electrostatic force acting on the contact line, obtained by integration of the pressure over a differential section of the surface is zero. In this situation, the electric pressure cannot contribute to the balance of forces on
the contact line; the local contact angle at the triple point remains unchanged and equal to Young’s value (Buehrle et al. 2003).

4.5. Application of Electrowetting

Although electrowetting has been explored by Lippman in 1875, substantial interest has been generated in its application recently in the field of MEMS and Microfluidics. The major application ranges from microdroplet manipulation for Lab on a chip to electrowetting displays and micropumps or microfluid dispensing tools.

4.5.4 Lab on a Chip

Lab on a chip application was conceived and promoted mainly by two laboratories, at Duke university under Richard Fair [71] and at UCLA by Kim [60][72] which was followed by different groups. The main idea was to create a series of electrodes, which can be programmed and activated electrically, and through a sequence of activation a small droplet of liquid can be moved on the surface of the electrode. The electrowetting technique was the most suitable technology for this cause but to conceive a device design the liquid must be in direct contact with the electrode.
A sandwich design, consisting of two parallel substrates was used, which later became a standard for micro fluidic actuation. One of the substrate contained programmable pattern electrodes which and the other substrate acted as a continuous ground, always in contact with the liquid droplet. The electrode made of a thin transparent ITO layer on a glass substrate is covered by a hydrophobic layer that gives rise to larger contact angle and less hysteresis but does not prevent electrical contact with the electrode. Kim et al [72] demonstrated that the liquid droplet can be transported, merged, mixed or splitted into two droplets through such a device. In order to allow reliable droplet actuation the droplets must overlap with at least two electrodes. So the smallest liquid droplet volume depends on the electrode size and for a substrate gap of
100-500 μm and electrode size of 1 mm the smallest volume of 0.1 μl – 1 μl was used for their experiments. After establishing the basic fluid manipulation techniques research is focused on reducing the activation voltages to approximately 20 V or less, so that a battery powered device for medical application can be designed. Another challenge faced was the biocompatibility of the materials and procedures as it was observed that the biomolecule adsorb un-specifically on the hydrophobic surfaces via hydrophobic interaction and electrostatic interaction. The adsorption was subdued by the use of oil instead of air as an ambient condition. Further applications like electrowetting assisted dip pen lithography, micro fluid dispenser and electrowetting driven valves were discussed by Mugele [46].

4.5.4 Micro Lenses

In mechanical equilibrium condition liquid vapor interface of a sessile droplet has a spherical shape. Depending on refractive index of the liquid properties light passing through the droplet is refracted and works as a spherical lens. Now application of electrowetting changes the curvature of the droplet, which in return changes the focal length of the lenses. These allows for the design of a flexible system with variable focal length, which can be controlled electrically. Berge [73] designed a closed cell filled with immiscible liquids, namely a nonpolar oil droplet within an aqueous salt solution and the lense operated without much hysteresis or degradation.
4.5.3 Liquid Paper

A very interesting electrowetting based application was proposed by Fenestra[74], which enables the concept of liquid display with potential use in the field of electronic paper. The concept is similar to the liquid lens where an oil droplet containing dissolved dye is confined between a square electrode and a top cover forming a pixel, surrounded by aqueous salt solution. At zero voltage the oil forms a continuous layer wetting the hydrophobic insulator at the bottom of the pixel, subsequently acting as absorbent of light and would not allow any reflection. When voltage is applied the oil film contracts and forms a droplet at the corner as a result covers only a fraction of the pixel. This gives rise to increase in reflectivity and can be easily used as on/off situation for pixels in a display.

4.5.4 Single Plate Electrowetting

Several groups have been working on the manipulation of micro droplets through the electrode array design. These electrode array designs require the droplet to be sandwiched between two planar surfaces or plates, one plate consist of patterned electrode array, a dielectric layer on it and a hydrophobic coating on top of it, the other plate consist of a continuous electrode plate, used as a ground, coated with a hydrophobic layer and supposed to be in contact with the droplet all the time. Cooney[75] reported single plate design for droplet manipulation where the upper plate is removed and the droplet is allowed to float on the electrode surface. A thin electrically conductive wire,
which resided between the dielectric and the hydrophobic layer, would work as a ground line. These ground lines are the substitute for the grounding function usually provided by the upper plate and the design is termed as grounding from below. The effect of the thickness of the dielectric and the width of the ground lines are observed and concluded

![Fig. 2 Electrowetting chips. Glass chip in the center of the circuit board carrier is the electrowetting chip. The gold ground lines are visible. Two arrays of electrodes with different grounding strategies are implemented on this chip. On the bottom section each electrode is surrounded by gold ground lines, whereas on the top section, each of the two columns of electrodes are surrounded by gold ground lines.](image)

Figure 4.6: Experimental setup of Single plate Electrowetting by Cooney [75]

that by decreasing the ground line width and/or increasing the dielectric thickness results in an increased contact angle change. Several cases of dielectric breakdown occurred and were attributed to the uniformity of the Teflon layer and also reaching the condition of maximum current passing through the liquid. The zero voltage contact angle for the Teflon layer was 112.7°. They proved theoretically and experimentally that the single plate design with ground electrodes perform well than the single plate design without
ground electrodes allows rapid movement of the droplet from one electrode to other and also does not slow down.

Figure 4.7: Manipulation of free droplets on a single planar surface by electrowetting method for mixing of two liquids [75].

The design of this single plate liquid manipulation device is an ideal design for our micromanipulation scheme. Our need to have a surface on the gripper is fully served by this design where a droplet can be manipulated electrically by using the electrowetting technique without using any extended wire of plate for grounding. The only difference is
that here in this design a sessile droplet has been considered and in our case a pendant
droplet manipulation is needed.

4.6 Experiments on Electrowetting

Electrowetting experiments require a layer of dielectric material which should
have the following properties according to Mugele et al. [46]

i) High dielectric constant

ii) Stable and chemically inert

iii) Large Young’s equation, hydrophobic

iv) Small contact angle hysteresis

One of the widely used materials that satisfy all these requirements is amorphous
Teflon AF®. This material can be spin coated or dip coated in the laboratory and a range
of thickness (tens of nanometer to few micrometers) can be achieved by varying the
process parameters. The contact angle of water on this surface is approximately 110° with
hysteresis in the range of 5° – 10°. When the experiments are performed in oil the contact
angle for zero degree was found to be 160° with less hysteresis. In [46] experiments were
performed with ac voltage (\(V_{\text{eff}} = 0 – 1 \text{kVolt}\) ) and frequency of \(v = 1-10 \text{ kHz}\) and contact
angle was found to be saturating near 30°.

In most studies a thick dielectric layer (> 10 μm) has been used, so to obtain
significant contact angle change the voltage requirement was greater than 200 V. It was
understood that the voltage requirements can be decreased by simply decreasing the
dielectric layer to nearly 1 μm or by applying a thin hydrophobic coating on top of the dielectric layer. Moon et al. [60] reported contact angle change of $120^\circ - 80^\circ$ with applied voltage as low as 15 V by using a hydrophobic fluoropolymer coating on top of the dielectric layer. The dielectric materials used in the experiments were 1 μm thick silicon dioxide, 12 μm thick parylene and 0.7 μm thick Barium Strontium titanate (BST) sample and the fluoropolymernr coating is a 1000 A coating of amorphous Teflon AF®. Their results indicate that thinner dielectric materials correspond to lower voltage requirements.

From the experiments performed by Verheijen & Prins [49] the contact angle modulation was found to be between $120^\circ$ to $60^\circ$ by application of voltage in the range of 0 V to 250V and 0 V to -250 V. Their experiments confirmed that electrowetting is independent of the polarity of applied voltage. The dielectric used was a 10 μm thick layer of parylene with a coating of 25 nm layer thick of AF1600.
CHAPTER V
SIMULATION OF DROPLET USING CFD ACE+

5.1 Introduction to Simulation Using CFD ACE+

CFD ACE+ is advanced simulation software capable of simulating multiple complex interacting physics (microfluidics, electrokinetics, biochemistry, electrostatics, stress etc.). This commercial multiphysics software has been used here to validate the steady state solutions obtained from solving Young-Laplace equation through the iteration method and to understand the transient characteristic of contact angle manipulation on droplet geometry change. The Volume of Fluid (VOF) module and Flow module had been used together for simulating the problem of liquid bridge behavior between two surfaces and investigating the effect of the change in contact angle on the meniscus geometry. The process of simulation has three steps

1. Pre-processing, a geometrical model was setup in CFD GEOM,
2. Simulation, the model is simulated with proper volume conditions and boundary conditions in CFD ACE module.
3. Post-processing, simulation results were visualized and analyzed in CFD VIEW module.
The first section of this chapter describes the theoretical background of the software. Then the results were discussed along with the validation of the software results with some experimental results. The validation process is completed and finally the numerical results from the iteration algorithm were matched with the CFD ACE results.

5.2 Free Surface Module (VOF)

CFD ACE+ utilizes a Free Surface Volume module which depends on a Volume of Fraction method to find out the interface between two immiscible liquids. This section is presents an overview of the module and the theories associated with it.

5.2.1 Introduction

Free Surface Module has the ability to model the fluid dynamics of two fluids, which are immiscible in nature and with high density ratios and produce the free surface geometry as a result. The effect of surface tension is taken into account for computation of the interfaces.

5.2.2 Free Surface Module (VOF) Theory

The theory for Free Surface Module (VOF) can be found in the reference manual of the software [78]. This Module uses the Volume-Of-Fluid (VOF) method based on the
works by Hirt and Nichols [79], and was recently extended by Rider and Kothe [80]. Their work emphasized on the Eulerian formulation for problems involving free boundaries, especially for free boundaries with large deformations where Langrangian methods cannot be used.

5.2.3 Free Surface Module Theory – Volume Fractions

The important feature of the VOF methodology is the definition of a parameter $F$ representing the fractional volume of fluid in a computational grid. i.e the fractal distribution of the second fluid (e.g. water) in the computational grid of primary fluid(air). Thus, $F$ takes the value 1 in cells that contain only secondary fluid (water) and the value 0 in cells that contain only primary fluid (air) and for a cell that contains an interface between these two fluids would have a value of $F$ between 0 and 1. For a given flow field and boundary conditions, $F$ can be determined by solving the transport equation defined as in (5.1)

$$\frac{\partial F}{\partial t} + \nabla \cdot \rho F = 0$$

$$\frac{\partial F}{\partial t} + \frac{\partial}{\partial x} (uF) + \frac{\partial}{\partial y} (vF) + \frac{\partial}{\partial z} (wF) = 0$$

where $F$ is the liquid volume fraction, $t$ is time, $\nabla$ is the standard spatial gradient operator, and $\nabla \cdot \nabla F$ is the velocity vector of the fluid. This equation must be solved together with the fundamental equations of conservation of mass and momentum (and energy when activated) in CFD-ACE+ to achieve computational coupling between the velocity field solution and the liquid distribution.
5.3 Flow Module Theory

The governing equations for flow of fluid in Flow Module are the general mathematical equations derived from the conservation laws of physics:

- Conservation of Mass. There is no loss or gain of mass in the system.
- Conservation of Momentum. The rate of change of momentum equals the sum of the forces on the fluid (Newton’s second law).

These two conservation equations form the Navier Stokes’s equation for flow and CFD-ACE+ employ an iterative method to solve for these equations.

5.3.1 Conservation of Mass

Conservation of mass requires that the time rate of change of mass moving out of a control volume be balanced by the net mass flow into the same control volume (outflow = inflow). This can be expressed as:

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \mathbf{V} = 0
\]

\[
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x} (\rho u) + \frac{\partial}{\partial y} (\rho v) + \frac{\partial}{\partial z} (\rho w) = 0
\]  \hspace{1cm} \text{5.2}

The first term on the left hand side is the time rate of change of the density (mass per unit volume). For incompressible flow the first term is zero. The second term describes the net mass flow across the control volume’s boundaries and is called the convective term. \( \mathbf{V} \) is
the velocity vector of the fluid flow which when transferred into $x$, $y$, $z$ direction can be written as $u$, $v$, and $w$.

5.3.2 Conservation of Momentum

Newton’s second law states that the time rate of change of the momentum of a fluid element is equal to the sum of the forces on the element. Conservation of momentum requires that the rate of change of momentum remains constant.

The $x$-component of the momentum equation is found by setting the rate of change of $x$-momentum of the fluid particle equal to the total force in the $x$-direction on the element due to surface stresses plus the rate of increase of $x$-momentum due to sources:

$$\frac{\partial (\rho u)}{\partial t} + \nabla \cdot (\rho V u) = \frac{\partial (- p + \tau_{xx})}{\partial x} + \frac{\partial \tau_{yx}}{\partial y} + \frac{\partial \tau_{zx}}{\partial z} + S_{MX}$$

Where $p$ is the static pressure and $\tau_{ij}$ is the viscous stress tensor and $S_{MX}$ is the Surface force in $x$ direction. When the viscous terms are neglected the steady state equation takes the form of Laplace equation. The surface tension forces are added through this term. Similar equations can be written for the $y$- and $z$-components of the momentum equation.

5.3.3 Navier-Stokes Equation

The momentum equations, given above, contain as unknowns, the viscous stress components $\tau_{ij}$, therefore a model must be provided to define the viscous stresses.
In Newtonian flows, the viscous stresses are proportional to the deformation rates of the fluid element. The nine viscous stress components (of which six are independent for isotropic fluids) can be related to velocity gradients to produce the following shear stress terms: The \( x \) terms are shown in Equation (5.4)

\[
\begin{align*}
\tau_{xx} &= 2\mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu \left( \nabla \cdot F \right) \\
\tau_{xy} &= \tau_{yx} = \mu \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)
\end{align*}
\]

5.4

Substituting these into the momentum equations gives the Navier-Stokes Equation for \( x \) component as shown in Equation (5.5)

\[
\begin{align*}
\frac{\partial (\rho u)}{\partial t} + \nabla \cdot (\rho u u) &= - \frac{\partial p}{\partial x} + \frac{\partial}{\partial x} \left[ 2\mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu \left( \nabla \cdot F \right) \right] + \\
\frac{\partial}{\partial y} \left[ \mu \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] + \frac{\partial}{\partial z} \left[ \mu \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} \right) \right] + S_{\text{xy}}
\end{align*}
\]

5.5

The details can be found in CFD ACE+ Modules Manual version 1[78]. See Numerical Methods for details on the methods used to solve these equations.

5.3.4 Theory of Mixture of Properties

From the Navier Stoke’s equation the question arises, what values of the parameters like density should be used for a cell containing the mixture of two fluids. The volume fraction \( F \) is used to determine the average density, viscosity, thermal conductivity, and specific heat capacity, etc. in a given computational cell. In particular,
the average value of any volume-specific quantity, in a computational cell can be computed from the value of $F$ in accordance with:

$$
\tilde{\phi} = F\phi_2 + (1 - F)\phi_1
$$

where $\tilde{\phi}$ is the volume-averaged quantity, $\phi_2$ is the value of the property for fluid two (water), $\phi_1$ is the value of the quantity for fluid one (air). For an intensive quantity, (5.6) must be extended to include the effect of density $\rho$ as shown in Equation (5.7).

$$
\tilde{\phi} = [F\rho_2\phi_2 + (1 - F)\rho_1\phi_1]/\rho_{\text{mix}}
$$

Fluids 1 and 2 can be interchanged. In principle, interchanging the fluids should have no effect on the results; in practice, it has a slight effect, which is more pronounced for problems with surface tension.

5.4 Numerical Scheme for Surface Reconstruction and CFL Number

CFD ACE+ employs a VOF method for tracking the interface of two liquid. It depends on the single scalar variable $F$ to describe the liquid distribution and solve for the liquid volume evolution. In this process the surface reconstruction is a pre requisite for determining the flux of the fluid 2 from one cell to the next cell and for determining the surface curvatures when the surface tension model is activated. It employs three methods of surface reconstruction

- A 0th order upwind scheme
- An upwind scheme with single line Interface construction (SLIC) method
An upwind scheme with the Piecewise Linear Interface Construction (PLIC) method

After computation of the interface the secondary fluid flux for a given velocity is determined by back projection of the cell face. Within each computational cell the stability limit is given by the so-called Courant condition

$$\Delta t_c = \frac{d_c}{|\vec{v}_c|}$$  \hspace{1cm} (5.8)

where $\Delta t_c$ is the maximum time step that can be taken in a cell $c$ where $\vec{v}_c$ and $d_c$ are respectively the velocity vector and local cell dimension (length scale).

The CFL (Courant, Freidricks, Levy) number is defined by

$$\left|\frac{\vec{v}_c}{d_c}\Delta t_c\right|$$  \hspace{1cm} (5.9)

The CFL number specifies the distance crossed by any phenomenon, effect or wave traveling at a velocity $\vec{v}_c$ as a fraction of local length scale of the cell. For example a CFL value of 0.2 would mean that the computation would allow the fluid interface to cross a maximum of 20% of the width of a cell during each time step. For stability the CFL number should satisfy the condition $\text{CFL} < 1$, but in practice values as low as 0.1 is often required. The value of the CFL number plays a crucial role in convergence of the problem and also for attaining the actual solution of the problem. For liquid interface simulations a CFL value of 0.2 has been recommended by the software manuals. We have conducted a study on the CFL Number which is produced in the later part of this chapter.
5.5 Limitations of Free Surface Module

The Free Surface Module has the following limitations:

- Must be run as a transient
- 3D hexagonal grid or 2D quads only for Surface Tension and higher order reconstruction (PLIC)
- Multi-to-one cell matching allowed (for example, 5 to 1 in 2D and 4 to 1 in 3D)
- Cannot solve for net charge on droplet
- Compatible only with the following modules:
  - Flow
  - Heat Transfer (no boiling)
  - Stress
  - Deformation
  - Electric and Magnetic Model (only Lorentz forces are fully accounted)
  - Chemistry (No gas phase chemistry. Only dilute liquid phase chemistry is allowed in FLUID ONE)
5.6.1 Grid Generation

The geometrical modeling is done with CFD GEOM module. Some important guidelines followed for the modeling is described below.

- A hexagonal structured grid in three dimensions and quadrilateral in two dimensions was strongly recommended for use with the Free Surface Module. Problems with surface tension require PLIC to compute curvatures and PLIC scheme requires a three-dimensional hexagonal or two-dimensional quadrilateral grid for efficient computation. The most ideal grid for a 2D VOF calculation is a square domain with square cells; the most ideal grid for a 3D VOF calculation is a cubic domain with cubic cells.

- It was advised to avoid performing any serial calculation with the VOF module using a grid with 100,000 or more cells. For simple problems the numbers of cells were recommended to be restricted below 10,000.

- For complex system or complex geometry, a component-by-component-modeling approach was highly recommended.

- It was mentioned that the solution quality highly depends on the grid-quality in general, and to the following aspects of it in particular:
  1. Poor Orthogonality affects flux calculations
  2. Poor smoothness aspect ratios above 10 affects the surface constriction

- Another important consideration affecting the design of a grid for a VOF computation is the number of cells required to resolve the shape of an interface. A
minimum of 4-5 cells are required across a gap (such as the gap between two flat plates, or the gap across the width of a channel) to provide adequate resolution of the shape of the interface in that gap. Using 8 cells across a gap provides ample resolution. If the necessary resolution is not available, the accuracy of the curvature calculation will be adversely affected.

5.6.2 Parameter Setup

In this section we will discuss some aspects of the simulation modeling and the choice of some parameters for problems specific to Liquid Bridge between two surfaces.

- A transient solution is needed for Free Surface Simulations. The Standard or Automatic Transient Option should be selected. For the Automatic time step option, input a target CFL number to limit the advection of the surface relative to the grid. For example, a CFL target of 0.2 (0.1 recommended) will limit the surface motion to 20 percent of the cell volume per time step.

- CFD-ACE+ provides two options in the VOF module: Surface Reconstruction and Time-Integration scheme.
  - There are three options available under Surface Reconstruction: 2nd Order (PLIC), 1st Order (SLIC), and 0th Order (None). The PLIC option should be selected to avail the Surface Tension and Pressure calculation options are available. The Surface Tension option must be selected to incorporate surface tension effects.
Two options are available for specifying the time-integration scheme, explicit and implicit. The explicit option corresponds to a Forward-Euler Scheme, in which the fluxes are computed from the $F$ distribution at the previous time-level. The implicit option corresponds to a Backward-Euler Scheme, in which the fluxes are computed from the $F$ distribution at the current time-level. Compared to the implicit option, the explicit option exhibits greater stability and better convergence, faster than the implicit option but slightly lower accuracy for a surface tension related problem.

- To assign boundary conditions on walls like contact angle, inlets, outlets and symmetry options the BC should be used. The VOF should be left as Fluid1 ($\text{Fraction} = 0$) to specify that the volume is occupied by the fluid 1 which is air and later the volume of the liquid 2, water can be specified as an initial condition for the solution.

- The Fluid 2 in sphere inside the geometry option suits better as an initial condition for droplet simulation. For some simulation the initial data is mapped from a previous solution using the appropriate option in IC.

- In the solver control section approximately 30 iterations are recommended per time step. We have used 50-100.

- One of the well-known defects of the VOF methodology, is the generation of tiny isolated droplets of liquid in gas regions, and of tiny isolated bubbles of gas in liquid regions. These droplets and bubbles are collectively called flotsam and jetsam. This effect becomes more pronounced with an increase in any of the following:
• Poor convergence, perhaps due to an insufficient number of iterations for each time step
• Too large a setting for the CFL Number
• Excessive skewness in the grid, or other contributors to poor grid quality.

The formation of flotsam and jetsam should be minimal with the PLIC surface reconstruction (compared to the SLIC surface reconstruction), but cannot be completely eliminated, especially if either the convergence or the grid quality is poor. The flotsam and jetsam filter attempts to eliminate flotsam and jetsam at the earliest stage of their formation, before they begin to adversely affect the solution, and before they grow in size. However, this filter should be regarded as a means of treating the symptom, and not the cause. Nevertheless, if the cause cannot be treated, then using this filter will at least prevent some of the negative side effects of flotsam and jetsam, not least of which is a reduction in the time-step size. The flotsam and jetsam filter can be turned on or off under the Advanced Tab in the Solver Control menu. The frequency, that must be supplied if the option is activated, should be set to 1.

- The capillary-wave damping option increases the viscosity in the vicinity of the interface, in an attempt to damp the capillary waves that are invariably generated in the interface under surface-tension forces.

The general guidelines for using capillary-wave damping are:

• Activate the capillary-wave damping option only if the interface motion is exhibiting symptoms of convergence problems.
• Use as little damping as possible to obtain the correct solution.
5.7 Simulation Results

CFD ACE+ is primarily used here to obtain the interface profile geometry of the liquid bridge between a spherical object and a planar surface. The numerical solutions obtained from the Young-Laplace equation are validated by the CFD ACE+ solutions. Our first step in this approach was to validate the simulation results with a real problem. The CFD ACE+ results were compared with an image of droplet. Next step was to find model the geometry and find a suitable mesh size for the simulations. At the end a number of simulation results are presented for different angles and different volumes.

5.7.1 Validation Of CFD ACE+ Results

We have selected the simplest real problem, shape of a droplet, for validation of the CFD ACE results. To achieve a comparison the shape of the interface of water and air was acquired from the digital image of a droplet. A micro syringe (company) was used to dispense 10μl of de-ionized water on a Teflon surface and the images were taken by a 6 mega pixel Canon S3 IS camera with super micro mode. The Teflon surface was cleaned with DI water and ethanol before the experiment and it was performed at room temperature 24°C and normal air pressure 76 mm Hg. The image is post processed in AutoCAD and from the dimensions and angles measured. The contact angle was found to be 96° as shown in Figure 5.1 (a).
Figure 5.1: (a) Digital image of 10 μl sessile droplet of water on a Teflon surface at room temperature 24°C. CFD ACE+ simulation results of a 10 μl sessile droplet with contact angle 96° for the conditions (b) with gravity and (c) without gravity.
The contact angle $96^\circ$ was used for CFD ACE+ simulation and the meniscus profiles obtained as shown in Figure 5.1(b) and 5.1(c). The height and the contact radius was calculated and the values presented in Table 5.1. The results were satisfactory close to each other. A relative error of 6.67% was recorded for simulation with gravity and 3.61% for simulation without gravity.

Table 5.1: The height and contact radius obtained from the simulation and measurement of the image of the droplet of volume 10 μl

<table>
<thead>
<tr>
<th>Type</th>
<th>height max D</th>
<th>height min D</th>
<th>height D</th>
<th>Contact radius max R₁</th>
<th>Contact radius min R₁</th>
<th>Contact radius R₁</th>
<th>contact angle θ₁</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>mm</td>
<td>mm</td>
<td>mm</td>
<td>mm</td>
<td>mm</td>
<td>mm</td>
<td></td>
</tr>
<tr>
<td>with g</td>
<td>2.06659</td>
<td>1.98275</td>
<td>2.02731</td>
<td>2.11638</td>
<td>2.03307</td>
<td>2.07716</td>
<td>96</td>
</tr>
<tr>
<td>without g</td>
<td>2.30343</td>
<td>2.19786</td>
<td>2.25085</td>
<td>2.00803</td>
<td>1.92638</td>
<td>1.96575</td>
<td>96</td>
</tr>
<tr>
<td>image</td>
<td>2.1724</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.10925</td>
<td>96</td>
</tr>
</tbody>
</table>

5.7.2 Effect of Mesh Size

This study was conducted to understand the dependence of computational time and accuracy on the mesh size. Same geometry was re-meshed by increasing the number of nodes and the simulation was run for 500 time steps for each case and the results were obtained for analysis. A total no of 8 cases were observed .It was found that the interface was not a discrete boundary and as the mesh size increases interface thickness reduces.
and the meniscus profile becomes more prominent and discrete. Meniscus thickness was thus taken as a measurement of accuracy of the simulation and was measured from the difference between minimum height and maximum height of the meniscus from the X axis. Table 5.2 shows the Interface thickness and computational time for 8 different mesh sizes.

Table 5.2 Comparison of computational time and interface thickness for different mesh size.

<table>
<thead>
<tr>
<th>Simulation</th>
<th>No of Quad Cell</th>
<th>Computational Time for 500 time step</th>
<th>Interface height $h_{\text{min}}$</th>
<th>Interface height $h_{\text{max}}$</th>
<th>Mean Interface thickness nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>sec</td>
<td>um</td>
<td>um</td>
<td>um</td>
<td>nm</td>
</tr>
<tr>
<td>1</td>
<td>1077</td>
<td>291.86</td>
<td>0.0815</td>
<td>0.1982</td>
<td>116.72</td>
</tr>
<tr>
<td>2</td>
<td>2404</td>
<td>705.06</td>
<td>0.1542</td>
<td>0.1960</td>
<td>41.84</td>
</tr>
<tr>
<td>3</td>
<td>3822</td>
<td>1192.84</td>
<td>0.1696</td>
<td>0.2015</td>
<td>31.93</td>
</tr>
<tr>
<td>4</td>
<td>5542</td>
<td>1659.77</td>
<td>0.1718</td>
<td>0.2004</td>
<td>28.63</td>
</tr>
<tr>
<td>5</td>
<td>8935</td>
<td>3204.84</td>
<td>0.1784</td>
<td>0.1949</td>
<td>16.52</td>
</tr>
<tr>
<td>6</td>
<td>10180</td>
<td>3746.16</td>
<td>0.1795</td>
<td>0.1949</td>
<td>15.42</td>
</tr>
<tr>
<td>7</td>
<td>10975</td>
<td>4189.69</td>
<td>0.1795</td>
<td>0.1971</td>
<td>17.62</td>
</tr>
<tr>
<td>8</td>
<td>13321</td>
<td>5099.03</td>
<td>0.1773</td>
<td>0.1949</td>
<td>17.62</td>
</tr>
</tbody>
</table>
Figure 5.2: Plot of computational time step and interface thickness when mesh size is increased from 1077 to 13321 quad cells.

Figure 5.3: Mesh geometry increased from (a) 1077 to (b) 13321 quad cells.
Figure 5.4: Interface geometry for different mesh size with no of quad cells (a) 1077 (b) 2404 (c) 3822 (d) 5542 (e) 8935 (f) 10180 (g) 10975 (h) 13321 quad cells.

Figure 5.2 clearly shows that if the mesh size is increased beyond 10,000 cells the computation time drastically increases but the interface thickness comes to a saturation point. This study helped us to concentrate on mesh sizes around 10,000 cells for any future simulations.
5.7.3 Contact Angle Manipulation For Volume 0.1 \( \mu \)L

The simulation procedure followed in this section is described below

1. Simulation is performed to obtain the geometry of a pendant droplet of volume 0.1 \( \mu \)l hanging from a surface with contact angle 60°. The height of the pendant droplet is calculated from the plot of the LIQVOF variable as shown in Figure 5.5. The variable changes value from 0 to 1 at the interface and the calculated mean values of \( h \) are taken as height of the droplet.

2. The height of the droplet will be considered as the gap between the spherical surface and flat surface. This model is parameterized and by changing the gap the CFD GEOM model can be created.

3. Now the simulation will be run for contact angle 60°. The initial condition for the water droplet is kept as a spherical droplet of volume 0.1 \( \mu \)l and contact angle 60°. A matlab code which solves for the center and radius of the circle when the gap, volume and contact angle is given has been used. It has been sometimes required to change the height or contact angle for proper start of the simulation and to overcome the problem associated with the bubble formation in the liquid bridge.

4. The output results of angle 60° were then used as initial condition for the next simulation with contact angle 80°. The simulations are performed up to 140° with interval of 20°. It was observed that at 140° the liquid bridge forms a necking and breaks down.
Figure 5.5: Calculation of height of pendant droplet of volume 0.1 μl with contact angle 60° from CFD ACE+ simulation results.

Figure 5.6 shows the simulation results for angle change of 60°-120°, and Figure 5.7 shows the simulation results corresponding to different time steps for contact angle 140°.

Figure 5.6: Meniscus profiles for 0.1 μl volume liquid bridge between spherical surface of radius 1 mm, contact angle 20° and a flat surface with contact angles (a) 60°.
Figure 5.6: Meniscus profiles for 0.1 μl volume liquid bridge between spherical surface of radius 1 mm, contact angle 20° and a flat surface with contact angles (b) 80° (c) 100° (d) 120°.(continued)
Figure 5.7: Simulation Results of meniscus profiles for 0.1 μl volume liquid bridge between spherical surface of radius 1 mm, contact angle 20 ° and a flat surface with contact angles 140° for time steps (a) 5275 (b) 6600 (c) 6800
Figure 5.7: Simulation Results of meniscus profiles for 0.1 μl volume liquid bridge between spherical surface of radius 1 mm, contact angle 20° and a flat surface with contact angles 140° for time steps (d) 7000 (e) 7513. (continued)

It should be noted that the simulation for 140° started with time step 5000 and ended at 7513 where the simulation was steady with two individual shapes of liquid. It can be inferred from the simulation that the liquid bridge ceases to exist when the contact angle is changes from 60° to 140°. The effect of electrical potential or electrostatic forces cannot be calculated from this simulation. The charge distribution on the liquid surface is also not available from this software. The meniscus profiles were extracted form the
results and used to compares with the results of the numerical simulation results as shown in Figure 5.8.

Figure 5.8: Comparison of solutions of meniscus profiles from CFD ACE+ analysis and numerical solution of Young-Laplace equation plotted in non dimensional x-y plane for angles (a) 60° and (b) 120°
5.8 Comparison of Results with Different CFL Number

We have conducted a study on the CFL numbers for liquid volume of 0.1 \( \mu l \), placed between a sphere of radius 1 mm and a flat surface, the contact angle of the spherical surface and the liquid is fixed at 20° and the contact angle of the flat surface and the liquid is changed from 60° to 140° in steps of 10°.

The CFL number was increased from 0.1 in increments of 0.1 and it was found that solutions exists up to 0.6 but the problem did not converge for values of 0.7 and above. The interface was plotted in MATLAB and the volume of the liquid bridge was computed to validate the convergence of the solutions.

Table 5.3 Comparison of volumes obtained from CFD ACE+ solutions with different CFL numbers. The actual volume for the simulation is 0.1 \( \mu l \) and \( \theta_2 = 20^\circ \)

<table>
<thead>
<tr>
<th>CFL Numbers</th>
<th>0.1</th>
<th>0.2</th>
<th>0.3</th>
<th>0.4</th>
<th>0.5</th>
<th>0.6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquid Volume ( \mu l ) ( \theta_1 = 60^\circ )</td>
<td>0.11087</td>
<td>0.11104</td>
<td>0.113398</td>
<td>0.113061</td>
<td>0.116013</td>
<td>0.115655</td>
</tr>
<tr>
<td>Liquid Volume ( \mu l ) ( \theta_1 = 120^\circ )</td>
<td>0.078747</td>
<td>0.066307</td>
<td>0.088490</td>
<td>0.093997</td>
<td>0.097523</td>
<td>0.094238</td>
</tr>
</tbody>
</table>

From this study it was found out that the for smaller contact angles the solution with smaller CFL number produces more accurate results but for higher contact angles the solution achieves higher accuracy within the range of 0.4 to 0.6. This gives a very valuable insight into the solutions achieved.
We can conclude that the CFL value plays a vital role in the accuracy of the solutions and for smaller contact angles a CFL number of 0.1 is best and for higher contact angles a CFL number of 0.5 is a suitable choice.

5.9 Comparison of Solution for No Slip Condition and Slip Condition

The CFD ACE+ solutions present the solution of Navier Stokes equations which is associated with velocity, shear force, pressure terms along with boundary conditions. The pressure term is formulated from curvature and surface tension terms. If the gravitational term is neglected and the solution is permitted to reach the steady state, the pressure terms should match with the solution of the Laplace equation. The Laplace equation on the other hand solves for the equilibrium equation corresponding to
minimum surface energy and relates the curvature with the pressure difference. Considering the differences in the problem formulation between these two methods, we conjecture that the effect of slip conditions on the boundary causes an inexact match between the solutions.

Figure 5.10: Comparison of solutions obtained from CFD ACE+ with no slip condition, slip condition on the surfaces and solutions obtained from ODE solution for (a) $\theta_1 = 60^\circ$ (b) $\theta_1 = 120^\circ$

Further simulations with CFD ACE+ have been performed by imposing slip condition to compare with the default no slip condition. It has been observed that for smaller contact angles imposing slip conditions do not alter the solution but for larger contact angles they do. This indicates that there might be a partial slip condition on the surface for higher contact angles.
5.10 Evaporation of Droplet

The experimental time for evaporation of 1 \( \mu l \) of sessile water droplet on hydrophobic surfaces was found to be in the range of 12-14 minutes in the following references. \([105][106].\) The evaporation process is fairly common but complex and evaporation time of the liquid bridge between two solid surfaces might be greater than the evaporation time of same volume of sessile droplet on a surface. The time for manipulation of a single object should be less than the time of evaporation of the droplet. For simplicity the effect of evaporation has been neglected in this study.

5.11 Effect on Capillary Number

The Capillary number represents the relative viscous forces with respect to the surface tension forces acting across an interface between a liquid and a gas or between two immiscible liquids. From the

\[
Ca = \frac{\mu v}{\gamma}
\]

where \( \mu \) is the viscosity of the liquid, \( v \) is the characteristic velocity and \( \gamma \) is the surface tension of the liquid in the gas phase. In this study we have been concentrating on validating the solutions obtained from Laplace equation by the results obtained from CFD ACE+. A steady state solution has been obtained in CFD ACE for that and as we are dealing with steady state solution it is considered that the Capillary number is zero for all the problems discussed here. However it was found that at higher contact angles the viscous terms does not die down to zero as seen for lower contact angle problems. This type of behavior of the CFD ACE+ solver needs to be studied in future.
5.12 Contact Angle Hysteresis

Contact angle hysteresis is another phenomenon which affects the contact angle when the liquid interface is moving on a solid surface. Advancing contact angle is greater than the receding contact angle on any surface. Hysteresis refers to the difference in these two contact angles. Walker et al [107] included the hysteresis effect in their Electrowetting modeling of EWOD surfaces. Due to change in advancing and receding contact angle the pressure at the interfaces differs, so they have scaled the pressure by a hysteresis constant which was obtained as a function of contact angles at zero voltage and angles at specified experimental voltage. The effect of contact angle hysteresis has been neglected in this scope of study. It is understood that the total hysteresis of the contact angle of water droplet on Teflon surface depends on the coating process and the surface roughness and can lie between $3^\circ$ and $27^\circ$. In this scope of study we have not modeled the electrowetting effect on contact angle and only the change in contact angle has been used to find out its effect on the stability of the liquid bridge. Hysteresis is an important parameter that should be dealt with in future study on this subject.

5.13 Conclusion

A commercial software CFD ACE+ was used to validate the results obtained from the solution of Young-Laplace equation. The simulation results from this software were first validated against the meniscus shape of a droplet of volume 10 $\mu$l by matching the simulation image to the digital image of the droplet. The results matched satisfactorily.
Then a number of simulations were performed where the contact angle was changed in step by step fashion and the effect of contact angle manipulation was observed. The results were matched with the results from numerical solution and it was found that the solution matches exactly for small contact angles, but for higher contact angles the solutions shows some discrepancy. The discrepancy can be attributed to the fact that Laplace equation does not take care of any viscous effect on the liquid bridge, but CFD ACE+ shows some viscosity effects at higher contact angles. As this software performs as transient solution the evolution of liquid meniscus shape with time was obtained. When contact is increased in step by step fashion a breakdown of liquid bridge was observed which also supports the concept of rupture as discussed in Chapter III before.
CHAPTER VI

STABILITY ANALYSIS OF AN AXISYMMTERIC LIQUID BRIDGE

6.1 Introduction

Capillary surfaces are governed by the nonlinear Young-Laplace equation as discussed in Chapter IV. In this study our focus is to understand the means to make a judgment that an equilibrium surface is stable or unstable. The history of the stability study is reviewed and important milestones covered in the following part of this chapter. Critical Stability study is performed in order to obtain valuable information on critical states of the parameters in the problem. This chapter will also give an insight on the amount of energy required to change any stable equilibrium state to unstable equilibrium states that would help us in breaking the liquid bridge easily at the time of release of the object.

The study of capillary surfaces dates back to Young (1805) [81] and Laplace (1806) [82] who formulated the basic concepts of surface tension. The study of the stability of bounded capillary surfaces began with Plateau (1863) [83]. He experimentally investigated capillary surfaces of a liquid bridge between two circular flat surfaces and was able to predict stability for special cases. As an historical aide Plateau [83] initially estimated the dimensionless critical length experimentally to be between 6 and 7.2 with
respect to the radius of the cylinder, but later he theoretically proved it to be exactly equal
to the circumference $2\pi$. Later Raleigh (1879) [47] clarified the matter theoretically and
determined the fastest growing wavelength was approximately nine times the radius of
the cylinder.

Howe (1887) [85] formulated the variational requirement for stability of
axisymmetric capillary surfaces in gravitation-less condition keeping the volume
constant. He solved for the Young-Laplace equation which is the first variation of the
energy functional and showed that the Legendre condition (Appendix C) for second
variation of the energy functional was always satisfied. Hence the stability was
determined solely by the absence of negative eigenvalues (a conjugate point in the Jacobi
equation). Howe’s approach of determining stability is termed as the classical stability
analysis and his work was continued by Myskhis (1987) [86] and Slobozhanin (1993)
[87]. Because of the limitations of the variational approach, such as inability to handle
fluid flow problem, various other approaches have also been used, as listed below:

1. Vega and Perales (1983) [88] showed that perturbation methods can be successful
   for a wide variety of problems, but the nonlinear nature of the problem limits their
   applications.

2. Meseguer Sanz, 1985[90] used Direct simulations with time dependent
   formulations.

3. Implicit numerical schemes by Martinez and Perales, 1986 [91].

Most of these methods require a separate complicated calculation of stability in addition
to equilibrium solution.
6.1.1 Classical Stability Analysis

The classical theory of stability analysis is best presented in a book named “Low Gravity Fluid Mechanics” authored by Myshkhis, Slobozhanin and their colleagues [86]. Their greatest contribution resides in development of the theory of stability for capillary surfaces and obtaining the solutions for several classical and applied stability problems. Based on the principal of minimum potential energy, the spectral stability criterion has been constructed by utilizing arbitrary perturbation of equilibrium surface and keeping the liquid volume constant. The spectral stability equations are basically eigenvalue problems and the smallest eigenvalues \( \lambda^* \) coincided with the minimum value of the \( \delta^2 U \), where \( U \) is the potential energy of the system defined later in this chapter. Consequently the equilibrium is stable if \( \lambda^* > 0 \) and unstable if \( \lambda^* < 0 \). Where \( \lambda^* \) is defined as \( \lambda^* = \min(\lambda_{01}, \lambda_{11}) \) where \( \lambda_{01} \) and \( \lambda_{02} \) are the smallest eigenvalues related to axisymmetric perturbations and the non-axisymmetric perturbations, respectively. The calculation of eigenvalues is a very complicated process so they proposed an effective method for understanding the stability by detecting critical axisymmetric states (\( \lambda^* = 0 \)) and constructing boundaries of the stability region. With this method Slobozhanin and his co-workers created different stability graphs for numerous problems like liquid bridges between equal disks in an axial gravity field [87], unequal disks in zero gravity condition [100], for rotating free mass of liquid for zero gravity [101], weightless liquid bridges [102]. The mathematics behind this scheme is described later in this chapter.
Their stability margin graphs mostly dealt with weightless liquid bridges between equal disks with free surfaces pinned to edges. A general boundary of the stability regions in \((\Lambda, V)\) plane was constructed where the disk slenderness is \(\Lambda\) and \(V\) is the relative fluid volume (ratio of the actual fluid volume to the volume of the cylinder pinned to disk edges). The stability margins which represents the height of a local potential energy barrier was determined for axisymmetric bridges that are linearly stable. If the total energy of the initial perturbation exceeds the stability margin, a considerable change in the equilibrium state (for example breakage of a bridge) should be expected.

6.1.2 Preferred Bifurcation Diagrams

An idea proposed by Padday and Pitt (1974) [92] and Boucher and Evans (1975) [93] described that stability may be deduced by observing the families of equilibrium curves without additional complicated calculation. This proposition was proved by Pitts (1976) [94] for axisymmetric drops subject to axisymmetric disturbances keeping the volume constant. In following years several authors noted the feature in limited context. Vogel (1989)[95] examined the case of axisymmetric liquid bridges with fixed contact angles and proved that turning points corresponds to stability changes for constant volume problems. Two factors limit the generalization of this approach to fixed contact line liquid bridges.
1. Liquid bridges of sufficiently high volume eventually becomes unstable due to non-axisymmetric disturbances, Plateau (1863)[83], Russo Steen (1986)[96], Slobozhanin and Perales (1993)[87].

2. Instability can occur at a branch point rather than a turning point (Boucher and Jones, 1988[97], Langbein 1992) [98].

In the applied mathematics community the development of bifurcation theory claims that the exchange of stability in turning points and at branch points is a general phenomenon. For the stability of equilibrium capillary surfaces the appropriate mathematical setting is the calculus of variation as shown by Laurey and Steen (1995) [99]. Their method delivers stability envelopes in the $LVB$ (Length, Bond Number, and Volume) parameter space for constant pressure and constant volume disturbances. They used preferred bifurcation diagrams (a plot of Volume $V$ and Pressure $P$), based on the properties of Jacobi equation, and claimed to produce stronger results than the classical bifurcation theory.

In this chapter the mathematical formulation of the spectral stability criterion has been discussed and the stability studies for the bridges were produced for understanding. The goal of this study is to set a framework to find a quantitative measurement of stability of the liquid bridge when the contact angle is manipulated. Most of the stability study dealt with a liquid bridge pinned at the edges of two planar surfaces. In our case, the problem is slightly different as the liquid bridge is formed between a planar surface and a spherical surface and the liquid bridge is not pinned to the edges of the surfaces but
is free to move with the change in contact angle. The stability study is performed with the focus that it could be used as a tool to predict the breakage of the liquid bridge.

6.2 Classical Stability Analysis of Equilibrium States

In general the stability of a mechanical system with finite number of degrees of freedom is determined by the sign of the second derivative of the continuous potential energy function of the system. For an equilibrium state to be stable the potential energy should be at its minimum state. When the solution of the parameters from the equilibrium equation (obtained by setting the first derivative of the energy equation with respect to the parameters to zero) is substituted in the second derivative, it should produce a positive value if the system is stable and negative if it is unstable. Now this stability criterion is true for rigid systems with finite number of degrees of freedom. Though it is natural to follow the minimum energy routine for stability, considerable difficulties arise as system concerned is in liquid state with free surfaces and with no degrees of freedom as defined in conventional sense. Myskhis [86] pioneered the method of calculation of stability margins by extending the concept of minimum energy for the state of liquid with free surface. The mathematical derivations discussed below can be found in his book. Lyapunov [86] first pointed out that the principle of minimum potential energy remains valid for equilibrium shapes of liquid if the stability definition is relaxed. Following his proposal, Rumyantsev [86] defined the stability criterion, and then Samsonov [86] modified his definition and proposed a theory for stability, which is described below. In
order to formulate the definition two parameters needed to be defined, \( l(\Gamma, \bar{\Gamma}) \), the
difference between the perturbed free surface \( \Gamma \) of the liquid and the unperturbed surface
\( \bar{\Gamma} \) and \( \rho(x, \bar{x}) \), the distance between points \( x \) and \( \bar{x} \) where \( x \) is position of a point on
the surface before perturbation and \( \bar{x} \) is the position after perturbation. The parameter
\( l(\Gamma, \bar{\Gamma}) \) is defined as the largest possible distance for two closest points as shown in (5.1)
\[
6.1 \quad l(\Gamma, \bar{\Gamma}) = \max_{x \in \Gamma} \min_{x \in \bar{\Gamma}} \rho(x, \bar{x})
\]
The largest possible acute angle between planes tangential to \( \bar{\Gamma} \) and \( \Gamma \) at the
corresponding points is denoted by \( \Delta(\Gamma, \bar{\Gamma}) \).

Definition:
The equilibrium shape \( \bar{\Gamma} \) of a liquid surface is called stable if for any positive \( L_1, L_2, L_3 \),
there is positive \( \lambda_1, \lambda_2 \) and \( \lambda_3 \) such that for any perturbed initial surface \( \Gamma_0 = \Gamma|_{t=0} \)
and initial velocity field \( v_0 \) satisfying the conditions
\[
6.1 \quad l(\Gamma_0, \bar{\Gamma}) < \lambda_1, \quad \Delta(\Gamma_0, \bar{\Gamma}) < \lambda_2, \quad |v_0| < \lambda_3
\]
The inequalities \( l(\Gamma_t, \bar{\Gamma}) < L_1, \quad E_t < L_3 \) are satisfied for all \( t \geq 0 \) or at least as long as
\( \Delta(\Gamma_t, \bar{\Gamma}) < L_2 \) where \( \Gamma_t \) and \( E_t \) perturbed surface and the kinetic energy of the liquid at
an instant \( t \).
The definition states that if any arbitrary small initial perturbation does not lead to significant deviation from its unperturbed state then it should be termed as stable. Though we have not followed this definition of stability it is to be noted that this definition is the basis for further stability theorems. Details can be found in the book, Low Gravity Fluid Mechanics pp 120-122[86].

6.2.1 Stability Theorem

So it is understood that for a certain position of absolute equilibrium of a liquid, if the second variation $\partial^2 U$ of the potential energy $U$ is positive then the position will be stable. Conversely if $\partial^2 U$ assumes negative values equilibrium position will be unstable.

Figure 6.1: Meniscus geometry, surface energies and contact angles at the contact line of liquid, gas and solid interface.
Now let’s define the potential energy $U$ for a liquid state with free surfaces by taking all the surface energy terms and the work done together as shown in (6.2)

$$U = \sigma|\Gamma| + \sigma_1|\Sigma| + \sigma_0|\Sigma_0| + \rho \int_{\Omega} \Pi d\Omega$$  \hspace{1cm} 6.2

where the parameters are defined as shown in Figure 6.1, $|\Gamma|$ is the surface area and $\sigma$ is the surface tension of the liquid-gas interface, $|\Sigma|$ is the surface area and $\sigma_1$ is the surface tension of the liquid-solid interface, $|\Sigma_0|$ is the surface area and $\sigma_0$ is the surface tension of the solid-gas interface, $\rho$ is the density of the liquid, $\Pi$ is the potential of a mass-force fields like gravitational forces on the volume of the system $\Omega$.

In order for a liquid in a vessel to be in equilibrium, the following hydrostatic conditions must be satisfied

a) Euler’s condition

$$\nabla p = \rho F \quad (\text{in } \Gamma)$$  \hspace{1cm} 6.3

where $p$ is the pressure in the liquid, $\rho$ is the density of the liquid and $\rho F(x)$ is the volume density of the forces on the surface $\Gamma$, where the mass-force field is assumed to have a potential $\Pi$, same as in (6.2) and the forces are derivative of the potential field

$$F = -\nabla \Pi$$  \hspace{1cm} 6.4

b) Laplace condition for the pressure drop at the interface between the liquid and the gas

$$p_0 - p = \sigma(k_1 + k_2)$$  \hspace{1cm} 6.5

Where $p_0$ is the constant pressure in the gas, $p$ is the pressure inside the liquid, while $k_1$ and $k_2$ are the curvature of the principal normal sections of the surface $\Gamma$. 
c) The Dupree–Young condition on the contact line of the liquid solid interface

\[ \sigma \cos \alpha = \sigma_0 - \sigma_1 \quad (\text{in } \gamma) \quad \text{(6.6)} \]

where the angle \( \alpha \) is the contact angle of liquid on a smooth surface as shown in Figure 6.1 and the equation is valid on the contact line \( \gamma \). In our case all the body forces like gravitational force, are neglected so we have concentrated on the Laplace equation (6.5) and Young’s equation (6.6).

6.2.2 Spectral Stability Criterion

For small perturbation \( (N \text{ is a perturbation function}) \) the expression for the second derivative of the energy equation (6.2) takes a form as shown below (pp.125, [86])

\[
\frac{1}{\sigma} \delta^2 U = \int (aN - \Delta N)Nd\Gamma + \int \left( \chi N + \frac{\partial N}{\partial e} \right)Nd\gamma
\]

(6.7)

Where the following notation has been used for sake of brevity

\[ a = \frac{\rho \frac{\partial \Pi}{\partial n} - k_1^2 - k_2^2}{\sigma} \quad (\text{on } \Gamma), \quad \chi = \frac{k \cos \alpha - \bar{k}}{\sin \alpha} \quad (\text{on } \gamma) \quad \text{(6.8)} \]

Where \( k \) and \( \bar{k} \) are curvatures of the normal cross sections of the surfaces \( \Gamma \) and \( \Sigma \) along \( \gamma \) and \( e_I \) respectively (pp. 105, [86]), while \( e \) are the unit vectors normal to \( \gamma \) in a plane tangential to \( \Gamma \) and \( e_I \) is the unit vectors normal to \( \gamma \) in a plane tangential to \( \Sigma \) as shown in Figure 6.1(a), while \( k_1 \) and \( k_2 \) are the curvature of the principal normal sections of the surface \( \Gamma \). \( \frac{\partial}{\partial e} \) is the derivative in \( \Gamma \) with respect to the outward normal to the \( \gamma \).

Here the perturbation function \( N \) must satisfy the condition of volume conservation.
According to the principles of Variational Calculus, the minimum of quadratic functional (6.7) under volume conservation condition (6.9) and the normalization condition (6.10)

$$\int_{\Gamma} N^2 d\Gamma = 1$$

6.10

corresponds to the smallest eigenvalue $\lambda = \lambda^*$ of (6.11). Therefore the corresponding normalized eigenfunction of the problem is

$$- \Delta N + aN + \mu = \lambda N \quad (\text{on } \Gamma), \quad \frac{\partial N}{\partial e} + \chi N = 0 \quad (\text{on } \gamma)$$

6.11

along with condition (6.9) as boundary condition.

This equation can be derived from the concept of Lagrange multiplier for minimization of a function $\frac{1}{\sigma} \delta^2 U$ with multiple constraints (6.9) and (6.10). $\lambda, \mu$ are the Lagrange multipliers whose values are not known beforehand. Combining (6.7), (6.9) and (6.10) one obtains

$$\int_{\Gamma} (aN - \Delta N)Nd\Gamma + \int_{\gamma} \left( \chi N + \frac{\partial N}{\partial e} \right) N d\gamma - \mu \int_{\Gamma} N d\Gamma + \lambda \int_{\Gamma} N^2 d\Gamma = 0$$

$$\int_{\Gamma} (aN - \Delta N - \mu + \lambda N)Nd\Gamma + \int_{\gamma} \left( \chi N + \frac{\partial N}{\partial e} \right) N d\gamma = 0$$

6.12

Further simplification of the above gives (6.12)

The sign of $\mu$ is ambiguous though. The constant $\mu$ and the orthogonality condition both can be eliminated by integrating both sides of the (6.11, first one). Then the eigenvalue problem assumes the form
\[-\Delta N + aN + \frac{1}{\Gamma} \int (\Delta N - aN) d\Gamma = \lambda N \quad (\text{on } \Gamma), \quad \frac{\partial N}{\partial e} + \chi N = 0 \quad (\text{on } \gamma)\]

6.13

Solving equation (6.11) should give the values of eigenvalues and eigenfunctions and the stability can be assessed by the sign of the minimum eigenvalue \(\lambda^*\) associated with both types of perturbations.

Condition of Equilibrium

1. If \(\lambda^* > 0\) the equilibrium state is **stable**
2. If \(\lambda^* < 0\) the equilibrium state is **unstable**
3. If \(\lambda^* = 0\) the equilibrium state is **critically stable**

Henceforth we shall speak of rough stability or rough instability depending on whether \(\lambda^* > 0\) or \(\lambda^* < 0\). The condition (6.8) is additionally required if \(\lambda^* = 0\).

For axisymmetric equilibrium surface \(\Gamma\), with cylindrical coordinates \(s, \theta\) the expression for \(\Delta N\) assumes the form

\[
\Delta N = \frac{\partial^2 N}{\partial s^2} + \frac{1}{r} \frac{dr}{ds} \frac{\partial N}{\partial s} + \frac{1}{r^2} \frac{\partial^2 N}{\partial \theta^2}
\]

6.14

Now if we take \(N(s, \theta)\) of the general form where the perturbation is a combination of axisymmetric and nonaxisymmetric terms as follows

\[
N = \varphi_0(s) + \sum_{n=1}^{\infty} \varphi_n(s) \cos n \theta
\]

6.15

For \(n = 1\), the value of \(N\) would have two parts, first part \(N = \varphi_0(s)\) corresponds to axisymmetric perturbation and the second part \(N = \varphi_1(s) \cos \theta\) corresponds to
nonaxisymmetric perturbation. Here it can be noted that the second term is a function of \( \theta \), which contributes to the nonaxisymmetric perturbation. Substituting (6.15) in (6.9) and (6.11) gives two sets of one dimensional boundary value problems corresponding these two terms in perturbation. The first equation is

\[
- \varphi_0'' - \frac{r'}{r} \varphi_0' + a(s) \varphi_0 + \mu = \lambda \varphi_0 \quad 0 \leq s \leq s_1
\]

with boundary conditions

\[
\left( - \varphi_0' + \chi \varphi_0 \right)_{s=0} = 0, \quad \left( \varphi_0' + \chi \varphi_0 \right)_{s=s_1} = 0, \quad \int_0^{s_1} r \varphi_0(s) ds = 0
\]

and second equation is

\[
- \varphi_1'' - \frac{r'}{r} \varphi_0' + \left( a(s) + \frac{n^2}{r^2} \right) \varphi_1 = \lambda \varphi_1 \quad 0 \leq s \leq s_1
\]

with boundary conditions

\[
\left( - \varphi_1' + \chi \varphi_1 \right)_{s=0} = 0, \quad \left( \varphi_1' + \chi \varphi_1 \right)_{s=s_1} = 0
\]

Here \( s \) is the length of the meniscus curve and it ranges from 0, initial contact point of the liquid and the solid surface, to the final point \( s_1 \) where the curve touches another solid surface. For all \( n \geq 1 \), the problem will be same as for \( \varphi_1(s) \), the only difference being that the coefficient \( \frac{n^2}{r^2} \) must be replaced in the corresponding equation by values of \( n \).

The corresponding eigenvalues takes the form \( \lambda_n = \lambda_1 + \frac{n^2}{r^2} \) (pp.133 [86]), so the eigenvalues increases as the value of \( n \) increases. As the stability criterion is dependent only on the minimum eigenvalues of the problem, the higher order perturbations can be
neglected and the stability analysis can be performed by considering only two types of
perturbation \( N = \varphi_y(s) \) and \( N = \varphi_z(s) \cos \vartheta \). Solution of (6.16) gives a set of
eigenvalues \( \lambda_{01} < \lambda_{02} < \lambda_{03} \ldots \) and solution of (6.18) gives a set of eigenvalues
\( \lambda_{11} < \lambda_{12} < \lambda_{13} \ldots \) for \( n = 1 \) and \( \lambda^* \) is defined as the minimum eigenvalue of the two set of
solutions \( \lambda^* = \min(\lambda_{01}, \lambda_{11}) \).

Once the perturbation equations are obtained the next step is to simplify the boundary
conditions. From the nature of dependence of \( \lambda^* \) on the parameters of the problem it can
be concluded that for higher values of \( a(.) \) and \( \chi \), \( \lambda^* \) will increase but is bounded (p.p
127 [86]) and for lower values of \( \chi \) \( \lim_{\chi \to -\infty} \lambda_{n1} = -\infty \). For a fixed \( \Gamma \) and a function \( a(.) \) with
upper bound it can be verified that \( \lambda^* \leq \nu^* \) where \( \nu^* \) is the lowest eigenvalue of a similar
problem with boundary condition \( N|_{\nu} = 0 \) and \( \nu^* \) does not depend on the value of \( \chi \).

So \( \lambda^* \leq \nu^* \) is always true for the above condition. Thus for \( \nu^* < 0 \), the equilibrium state
of the liquid is always unstable. On the other hand if \( \nu^* > 0 \) by a suitable choice of
curvature we can make it stable (if \( -k \) is quite large) or unstable (if \( k \) is quite large)
without changing the equilibrium surface \( \Gamma \). So now the problem can be redefined with a
new set of boundary conditions.

For a given force field and given integral line the values of minimum eigen values of
(6.16) and (6.18) \( \lambda_{01} \) and \( \lambda_{11} \) continuously depends on \( s \), and \( \chi_1 = \chi|_{s=s_1} \), increasing with
\( \chi_1 \) as shown in (6.20)
\[ \lim_{z_1 \to -\infty} \lambda_{n_1}^1 = -\infty, \quad \lim_{x_1 \to -\infty} \lambda_{n_1}^1 = v_{n_1} \quad (n = 0, 1) \quad 6.20 \]

Where \( v_{n_1} \) is the smallest eigenvalues \( v_{n_1} \) of analogous problems with boundary conditions \( \phi(s_1) = 0 \). The values \( v_{n_1} \) are continuous decreasing function of \( s_1 \) and 

\[ \lim_{s \to 0} v_{n_1} = \infty, \]

Summarizing the problem we can write that for perturbation function of the form in (6.18) where one term of axisymmetric perturbation and first term from nonaxisymmetric perturbation is considered

\[ N = \phi_0(s) + \phi_1(s) \cos \theta \quad 6.21 \]

then according to Myshkis [86] the stability of the axisymmetric equilibrium liquid bridge can be reduced to the determination of the sign of the smallest eigenvalue \( v^* = \min(\nu_{01}, \nu_{11}) \) of a known spectral problem. The spectral problems (6.16 - 6.19) can be reduced to one dimensional boundary value problems and \( \nu_{01}, \nu_{11} \) are the smallest eigenvalues of the following problems

\[ L\phi_0 - \mu = \nu_0 \phi_0, \quad 0 \leq s \leq \nu_1 \quad 6.22 \]

\[ \phi_0(0) = 0, \quad \phi_0(\nu_1) = 0, \quad \int_0^{\nu_1} r \phi_0(s) ds = 0 \]

where \( \mu \) is a constant and the eigenvalues of this problem are continuously increasing i.e. \( \nu_{01} < \nu_{02} < \nu_{03} \ldots \) associated with the axisymmetric perturbation. One dimensional boundary value problem associated with nonaxisymmetric perturbation is
and the eigenvalues of this problem are continuously increasing \((v_{11} < v_{12} < v_{13} \ldots)\). Here the operators are defined as

\[
L \varphi + \frac{1}{r^2} \varphi = v \varphi, \quad 0 \leq s \leq s_1
\]

\[
\varphi_1(0) = 0, \quad \varphi_1(s_1) = 0,
\]

and prime denotes derivative with respect to \(s\), where \(s\) is the arc length of the liquid bridge meniscus and \(\beta\) is curvature of the meniscus and \(B\) is Bond Number defined as

\[
B = \frac{\rho g R^2}{\sigma}
\]

where \(R\) is a characteristic length, \(\rho\) is density of the liquid and \(\sigma\) is the surface tension of liquid gas interface. This complicated form of equation is difficult to solve so another approach of finding out the critical stability criterion has gained popularity.

The critical stability criterion can be obtained by setting the eigenvalues of the problems to zero, i.e. \(v_{01} = 0\) \((v_{11} = 0\)). The method of determining the critical axisymmetric equilibrium state was described in [86]. It’s application to the stability problem of the liquid bridges between equal disks and in an axial gravity field of weightless liquid bridges was demonstrated in [87] and [84].
6.2.3 Critical Stability Criterion

The problem of critical stability can be solved by solving the equations (6.22) and (6.23) with eigenvalues \( \nu \) set to zero [86]. The presence of the constant term \( \mu \) in equation (6.22) makes it complicated to solve. So equation (6.22) is not solved directly and the solution of (6.22), \( \varphi_0 \) is considered to be a linear combination of the two different solutions \( \varphi_{01} \) and \( \varphi_{02} \) of equations (6.26) and (6.27), i.e. \( \varphi_0 = c_1 \varphi_{01} + \mu \varphi_{02} \) where \( c_1 \) and \( \mu \) are unknown constants.

The vanishing of any eigenvalues of the problem (6.22) at the boundary point \( s_i \) is equivalent to the fact that some nontrivial solution \( c_1 \varphi_{01} (s) + \mu \varphi_{02} (s) \) satisfies the boundary conditions of (6.22),

\[
c_1 \varphi_{01} (s_i) + \mu \varphi_{02} (s_i) = 0, \quad c_1 \int_0^{s_i} r \varphi_0 (s) ds + \mu \int_0^{s_i} r \varphi_0 (s) ds = 0 \tag{6.25}
\]

which is equivalent to the equality \( D(s_i) = 0 \) where \( D(s) \) is defined by taking the determinant of the boundary conditions of (6.22)

\[
D(s) = \varphi_{01} (s) \int_0^s R \varphi_{02} (s) ds - \varphi_{02} (s) \int_0^s R \varphi_{01} (s) ds \tag{6.26}
\]

and \( \varphi_{01}, \varphi_{02} \) are the solutions of two equations ,

\[
L \varphi_{01} = 0 \tag{6.27}
\]

With boundary conditions \( \varphi_{01} (0) = 0, \ \varphi_{01}' (0) = 1 \) and

\[
L \varphi_{02} = 1 = 0 \tag{6.28}
\]
with boundary conditions \( \varphi_{02}(0) = 0, \quad \varphi'_{02}(0) = 1 \)

The vanishing of any eigenvalues of the equation (6.23) is equivalent to the equalities \( \varphi_{11}(s_1) = 0 \), where \( \varphi_{11}(s) \) is solution of the equation (6.29)

\[
L \varphi_{11} - \frac{1}{R^2} \varphi_{11} = 0
\]

with boundary conditions \( \varphi_{11}(0) = 0, \quad \varphi'_{11}(0) = 1 \).

![Diagram showing stable, critically stable, and unstable meniscus profiles for axisymmetric liquid bridge](image)

Figure 6.2: Position of the critically stable points for stable, critically stable and unstable meniscus profiles for axisymmetric liquid bridge.

From the properties of \( \nu_{01} \) and \( \nu_{11} \) it follows that the liquid bridge is stable with respect to axisymmetric (or nonaxisymmetric) perturbations if the function \( D(s) \) (or the function \( \varphi_{11}(s) \)) does not vanish for the range \( 0 \leq s \leq s_1 \) where the length of the
meniscus profile \( s \) ranges from 0 to \( s_1 \). The first point where \( D(s) \) or \( \varphi_{11}(s) \) vanishes (in practice changes sign) is critical point \( s = s^* \). As shown in Figure 6.2 if \( s^* > s_1 \) then axisymmetric equilibrium state is stable, if \( 0 \leq s^* \leq s_1 \) then it is unstable. The profile for which \( s_1 = s^* \) corresponds to critical stable equilibrium state.

This procedure avoids the calculation of complicated equilibrium problem and facilitates the determination of critically stable equilibrium surface. The typical critical stability graph which looks like Fig 6.1 [87]. The critical values of the parameters are solved by tracking the change in sign of \( D(s_1) \) and the function \( \varphi_{11}(s_1) \) for axisymmetric instability and non axisymmetric instability. The parameters for the graph are dimensionless slenderness and dimensionless relative volume which are defined as

\[
\Lambda = \frac{d}{2r_0} \quad V_n = \frac{V_{\text{actual}}}{\pi r_0^2 d}
\]

Where \( r_0 = \frac{r_1 + r_2}{2} \), \( r_1 \) is the contact radius on the plane surface and \( r_2 \) is the contact radius on the spherical surface.

To find the critically stable surfaces an iteration method was followed. For their specific problem of a liquid bridge between two circular flat surfaces with equal radius \( (r_0) \) [102], where the liquid is pinned at the edges, the bond number was controlled by changing the contact radius \( (r_0) \). For a constant \( r_0 \), and constant initial contact angle \( \beta_0 \), the Laplace equation was solved iteratively to find out the shape of the meniscus and equations (6.25-6.28) are solved to find the values of \( D(s_1) \) and the \( \varphi_{11}(s_1) \) where one of them becomes zero. As the solution depends on pressure difference, iteration was
performed on pressure difference until a solution is found for which the liquid meniscus touches the edge of the upper plate. This solution provides a critically stable point on the stability graph. Other points can be determined by changing the initial contact angle $\beta_0$. Different sets of graphs can also be obtained by changing the Bond number, i.e. contact radius $r_1$. By a dense system of graphs a region in the stability graph is found out inside which all the solutions reside. To understand this graph we will discuss the stability margin for stable weightless liquid bridges as Slobozhanin described in his paper [102]. Figure 6.2 is created from the data available in [102].

![Stability Diagram](image)

**Figure 6.3: Generic stability diagram for axisymmetric weightless liquid bridges [102]**

The boundary of the stability region consists of three main branches, Am, CDEFn and ABC as shown in Figure 6.3. At the upper branch $Am$, the loss of stability occurs due
to nonaxisymmetric perturbations, the surfaces of critical bridges are rotund nodoids with convex profiles and with contact angles $\beta_1 = 0^\circ$ and $\beta_2 = 180^\circ$. At the branch $CDEFn$ the stability is lost due to axisymmetric perturbations. This surface has two distinct parts. The critical points corresponding to the segment $EFn$ are rotund (within $Fn$) or constricted unduloids (within $EF$) with contact angles constant at $90^\circ$. At the point $F$ ($V = 1, \Lambda = \pi$), the critical surface takes a cylindrical shape. For the segment $CDE$, the critical boundary curve is a locus of the turning points in volume where it reaches the minimum value, and the contact angles lies in the range $90^\circ < \beta_1 < 180^\circ$. The shapes are constricted unduloids within the segment $DE$, a catenoid at the point $D$, and constricted nodoids within $CD$. Finally, the left-hand segment $ABC$ of the lower boundary corresponds to instability occurring due to detachment of liquid bridge from the top disk. The shapes are constricted nodoids that have concave profiles with contact angles $\beta_1 = 180^\circ$ and $\beta_2 = 0^\circ$. The figure contains some bifurcation points as $T_1, T_2, T_3$ as the bifurcation structure is of particular importance for the stability margin problem. It was found that bifurcation is subcritical along the boundary segments $T_1AT_2$ and $T_3CDEFn$, and is supercritical (i.e. out from stability region) along the segments $T_1m$ and $T_2BT_3$. A small note here is the contact angle defined in this figure is exactly the opposite way we have defined our contact angle, i.e. our definition for $\beta_1 = 180^\circ$ is defined as $\beta_1 = 0^\circ$ in [102]. It should also be noted that these curves corresponds to Bond Number 0 and any plot corresponding to finite Bond Number value lies inside this region.
Most of the available graphs are for capillary bridge between two circular flat surfaces, where the liquid is pinned at the edges. The interesting part of this system is that when the volume is changed the contact angle at the edges changes. In our problem we are more interested in a liquid bridge between a sphere and a plate and the liquid edges are not pinned on the edges, as a result when the volume is changed the contact edges moves along the surface keeping the contact angles fixed.

In order to find out the boundaries of the stability graph in our problem we solved for the Young Laplace equation and critical stability criterion (6.25-6.28) by numerical integration method. For a constant value of \( r_1 \) and constant initial contact angle \( \theta_1 \) the equations were solved to find out the critical points \( s^* \) where either the value of \( D(s_1) \) or the value of \( \varphi_{11}(s_1) \) vanishes. Vanishing of \( D(s_1) \) and \( \varphi_{11}(s_1) \) corresponds to critical stability point due to axisymmetric perturbation and nonaxisymmetric perturbation respectively. For a certain value of \( r_1 \) a number of critical points can be obtained by changing the pressure difference \( \Delta p \). For the same contact angle \( \theta_1 \) when the contact radius \( r_1 \) is changed another curve is obtained. All these curves are plotted on a preferred stability coordinates by calculating the parameters \( V \) and \( \Lambda \). When the simulation is run for different values of contact angle \( \theta_1 \) a set of curves can be found. A stability margin can be obtained by selecting the minimum volume points for each curve corresponding to angle \( \theta_1 \). Figure 6.3 shows the stability boundary curve.
Figure 6.4: Critical Stability diagram for axis-symmetric weightless liquid bridges between a sphere and a plate for different contact angle \( \theta_1 \):

- (a) \( \theta_1 = 0^\circ \),
- (b) \( \theta_1 = 10^\circ \),
- (c) \( \theta_1 = 30^\circ \)
Figure 6.4: Critical Stability diagram for axis-symmetric weightless liquid bridges between a sphere and a plate for different contact angle $\theta_1$ (d) $\theta_1 = 90^\circ$, (e) $\theta_1 = 120^\circ$ (continued).
Figure 6.5: Critical Stability boundary for axis-symmetric weightless liquid bridges between a sphere and a plate.

Observations

1. All the points on the curve in Figure 6.4 and Figure 6.5 are critically stable points corresponding to axisymmetric or nonaxisymmetric perturbation.

2. All the plots in Figure 6.4 are plots for different values of $r_1$; the curves are formed by changing values of $\Delta p$. The range of $\Delta p$ is chosen in such a way that it covers all the possible solutions.
3. For a contact angle $\theta_1$ ranging from $0^\circ$ to $90^\circ$, Figure 6.4 (a), (b), (c), and (d) follows the same pattern for different values of $r_1$ and is very close to each other. For angle $120^\circ$ the curves deviates for different $r_1$ values.

4. For each angle the curve with the maximum slenderness point is chosen and the minimum volume point from that curve is taken as the boundary values for that angle $\theta_1$. These minimum volume points for each curve, B, C, D, and E, along with two extreme points A and N were used to create the stability boundary curve. The concept was to form a stability boundary consisting of critical points as the stability boundaries created in the literature corresponds to the critical points.

5. The segment EN is critically stable line due to nonaxisymmetric perturbation. Segment AE is critical to axisymmetric perturbation

Simulation study of the evolution of meniscus profiles by changing the gaps for contact angle $\theta_1 = 60^\circ$ and $\theta_2 = 20^\circ$ was performed for different constant volumes. The data was plotted in Figure 6.6 with the stability boundary obtained from the Figure 6.5 to find correlation between the two data
Figure 6.6: Stability instability curves for constant volume simulations for varying gaps $D$, volumes used were 1 µl, 0.1 µl and 0.01µl. The stability boundary obtained from Figure 6.5. Solid lines are stable solutions and dashed lines unstable solutions

Observations

1. The stable and unstable solutions obtained by manipulation of gap $D$ for a constant volume, constant contact angle $\theta_i$, was plotted in Figure 6.6. It was noticed that the stability and instability curves converges to a single point. This converging point is the critical point and it was observed that they lie close to the critical stability boundary curve obtained from Figure 6.5.
Simulation study of the evolution of meniscus profiles for changes in contact angle $\theta_1$ with constant gap $D$ and contact angle $\theta_2=20^\circ$ was performed for different constant volumes. The data was plotted in Figure 6.7 with the stability boundary obtained from the Figure 6.5 to find correlation between the two data.

![Graph showing stability boundary and varying contact angles](image)

Figure 6.7: Stability instability curves by varying contact angles $\theta_1$ for constant volume operations. Volumes used were 1 $\mu l$, 0.1 $\mu l$, 0.01 $\mu l$, and 0.001 $\mu l$. The stability boundary obtained from Figure 6.5. Solid lines are stable solutions and dashed lines unstable solutions.
Observation

1. For contact angle manipulation it was observed that the stable and unstable solutions converge to a point when the contact angle $\theta$ is increased from $60^\circ$ to $150^\circ$. It was found that between $144^\circ$ and $148^\circ$ there lays a critical contact angle after which there is no feasible solution.

2. The stability boundary obtained from Figure 6.5 does justify the manipulation of gap but fails to provide adequate information to justify this behavior of contact angle manipulation. Also it was observed that the location of the converging points for volumes 1 $\mu l$ and 0.1 $\mu l$ cannot be obtained precisely.

6.3 Preferred Diagrams

In this section we will discuss about the application of preferred diagrams for stability analysis. Following the paper from Lowry and Steen [99] the stability can be studied from preferred diagrams without getting into complicated stability analysis calculations. Maddock’s theorem has been used to justify the stability analysis by using preferred diagrams. Maddock’s theorem considers minimization problems of the form (6.31)

$$F[u] = G[u] - \lambda H[u]$$

where $F[u]$ is a functional and the objective is to minimize $G[u]$ subject to $H[u]$ prescribed (the constraint problem] or subject to $\lambda$ prescribed, the unconstrained problem. Maddock’s theorem demonstrates that the stability and instability states for both the
constrained and unconstrained cases can be determined by the equilibrium solutions in the preferred bifurcation coordinates of $H$ against $\lambda$. Turning points on these graphs always corresponds to change in sign of a single eigenvalue. Two distinct types of turning point can be observed, one corresponds to gain in stability, where unstable solutions changes to stable solutions and one for loss of stability where stable solutions becomes unstable by change in bifurcation parameters. From the bifurcation theorem it is known that if a brunch of stable states undergoes a turning point plotted as dependent variable against a bifurcation parameter, then there is a change in stability at the turning point.

The problem of general capillary surfaces like pendant drops with constant contact line pinned at the edges or constant contact angle, with volume preserved, falls within the same form of (6.31), where $G[u]$ is the energy functional, sum of the surface and potential filed energies to be minimized with keeping the volume constant, $H[u]$ is the functional corresponding to volume constraint. It has been proved before that for a capillary liquid bridge problem the parameter $\lambda$ accounts for the pressure jump across the capillary surface $\Delta p$. So the coordinates Volume and Pressure are preferred coordinates under this general framework. Different forms of liquid bridges between two circular flat surfaces have been studied in [99] and preferred diagrams were obtained to show that the turning points in PV diagram describe the exchange of stability and instabilities. A note here, the preferred coordinates are taken as the scaled volume, ratio of actual volume to volume of a cylinder with mean radius and gap and pressure inside the liquid.
Incorporating this theorem to our problem we studied the preferred diagrams for change in gap and change in angle and the observations are presented below in Figure 6.8 and Figure 6.9.

Figure 6.8: Plot of equilibrium solutions in the preferred coordinates $V_n$ and $\Delta p$ for constant volumes operations where the gap $D$ is being changed. Solid lines are stable solutions and dashed lines unstable solutions.

Observations

1. As indicated by Maddock’s theorem[99] for constrained problems, where the actual volume is fixed, as the bifurcation parameter $\lambda$ is increased in the preferred
coordinates $H-\lambda$ if the fold opens upwards, the number of negative eigen values changes from $j$ to $j+1$ and if the fold opens downwards the number of negative eigen values changes from $j$ to $j-1$. Which means stability is lost at a turning point in the preferred diagrams and the turning points are the critical points corresponding to the critical stability analysis where the eigenvalues changes sign.

2. Figure 6.8 demonstrates the Maddocks theorem as it can be seen that the plots for all the constant volume operations in the preferred plot have a turning point where stable solutions changes into unstable solutions.

Now the equilibrium solutions corresponding to contact angle manipulation is plotted in Figure 6.9
Observations

1. When the equilibrium states for change in contact angle $\theta_1$ is plotted on the P-V coordinates it can be observed that there are no turning points on the plot where the stability changes to instability, instead stability instability curves seem to be continuous and critical points lies in between.

From [99] it was learnt that Maddock showed using a counterexample that eigenvalue/turning point relationship is not true for preferred diagrams in general. The
same rules for sign of eigenvalue change in preferred coordinates can be applied to any other diagram by simple changes in preferred coordinates. The statement that draws attention is that the preferred diagrams are not unique in its properties and turning points may be visible if another set of preferred coordinates can be defined.

In our specific case for contact angle manipulation we fail to observe any turning point for the preferred coordinates $V$ and $P$. So new preferred parameters were defined and preferred diagrams plotted where turning points were obtained. The new parameters are contact radius ratio $K$, scaled contact angle, and contact angle ratio defined as in 6.32.

$$K = \frac{r_1}{r_2} \quad \Phi = \frac{\theta_1}{r_0} \quad \Theta = \frac{\theta_1 + \theta_2}{2} \quad \Psi = \frac{\theta_1}{\theta_2}$$  \hspace{1cm} 6.32

Figure 6.10 is the plot of the equilibrium points on the preferred coordinates $K$ and contact angle $\theta_l$. 
Figure 6.10: Plot of equilibrium solutions in the preferred coordinates $K$ (contact radius ratio $r_1/r_2$) and contact angle $\theta_1$ for constant volumes operations where the contact angle $\theta_l$ is being changed. Solid lines are stable solutions and dashed lines are unstable solutions.

Observations

1. It was observed that for all the volumes the critical contact angle lies in the range of 144°-148°.

2. The parameter $K$ reaches a maximum value at the critical point, which makes sense as the ratio of $r_1$ and $r_2$ increase the liquid bridge has maximum contact area on one surface and minimum contact area on the other surface, which means the liquid is leaving one surface and moving onto the other surface.
Figure 6.11: Plot of equilibrium solutions in the preferred coordinates $V_n$ (defined as $V_{actual}/V_{cyl}$) and scaled contact angle $\theta_i/r_0$ for constant volumes operations where the contact angle $\theta_i$ is being changed. Solid lines are stable solutions and dashed lines unstable solutions.

Observations

1. The critical point for the scaled parameters increases with decreasing volume which is evident because it was already noted that the critical contact angles are
very close to each other for any volume and for less volume the mean contact radius decreases.

Figure 6.12: Plot of equilibrium solutions in the preferred coordinates $V_n$ (defined as $V_{\text{actual}}/V_{\text{cyl}}$) and mean contact angle $\Theta$ for constant volumes operations where the contact angle $\theta_i$ is being changed. Solid lines are stable solutions and dashed lines unstable solutions.
Figure 6.13: Plot of equilibrium solutions in the preferred coordinates $V_n$ (defined as $V_{\text{actual}}/V_{\text{cyl}}$) and contact angle ratio $\Psi$ for constant volumes operations where the contact angle $\theta_1$ is being changed. Solid lines are stable solutions and dashed lines unstable solutions.

6.4 Stability Margin

For a stable equilibrium state of a mechanical system the stability margin, $M$, can be defined as the height of a local potential wall (or depth of a potential well) or the energy required to jump from a stability point to an unstable point. In terms of energy it is defined by 6.33.
where $U_e$ is the value of the potential energy at the point of emergence from the well, and $U_s$ is the stable equilibrium state corresponding to the value of potential energy at the bottom of the well. Myskhis pioneered the work on stability margin for equilibrium states of capillary liquid. His finding was that, this approach based on the equation 6.33 is possible if an unstable equilibrium state determining the point $U_e$ can be detected and a bifurcation study at the critical region proves that the bifurcation is subcritical. A bifurcation is subcritical when it takes place towards the stability region, which means that for some values of the bifurcation parameters around the critical region there are two equilibrium states, one stable and another unstable state which bifurcates in the same direction from the critical point. In that case the stability margin can be calculated by finding the difference between potential energy of the unstable state and the stable states. Though numerous studies on different problems have been performed, no systematic solution for the stability margin problems have been found yet. Among the previous works, the study of stable axis-symmetric states of weightless liquid bridges, pinned to edges of two coaxial, circular flat disk of same diameter by Slobozhanin [102] is notable. In light of this approach, surface energy terms were calculated and plotted for our problem for two different cases, the gap change and the contact angle change.
Figure 6.14: Surface Energy plot for stable unstable equilibrium solutions with respect to dimensionless Gap $D$ for constant volume operation with volumes (a) 1 $\mu l$ and 0.1 $\mu l$, (b) 0.01 $\mu l$ and 0.001 $\mu l$. 

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The plots of Surface Energy are presented in Figures 6.14-6.15. Figure 6.14 shows the surface energies of the stable and unstable states for constant volume operations when the gap is increased up to the point of no solution.

Observations

1. For each plot of constant volume curves in Figure 6.14 it can be seen that for small gaps there are only one solution, when the gap is increased the unstable solutions appear. The surface energy associated with the stable solutions continuously increases and at a critical gap the two solutions coincides. After the critical gap is reached there are a few occasions when a solution is found, but as the gap is increased further, no solution can be obtained.

2. The unstable solutions occur on the same side of stable solutions with respect to the critical point so it can be concluded that the bifurcation is subcritical for this problem. For any gap where there are two solutions, the difference in surface energies can be termed as the stability margin for that gap.

3. The physical meaning of the stability margin is that, if a liquid bridge is in its stable state and we know the stability margin for that state then by application of energy greater than the stability margin, from outside of the system, one can disrupt the stability of the state.

4. The critical gap is dependent on the volume of the liquid. The order of stability margin decreases with decreasing volume. A quantitative analysis of energy associated with free vibration of a gripper would give us an idea, what volume is
needed for a stable manipulation. For a stable manipulation the stability margin should not be less the energy associated with the natural disturbances on the system.
Figure 6.15 Surface Energy plot for stable unstable equilibrium solutions with respect to dimensionless contact angle $\theta_1$ for constant volume operation with volumes (a) 1 $\mu l$ and 0.1 $\mu l$, (b) 0.01 $\mu l$ and 0.001 $\mu l$. 
Observations

1. For the contact angle manipulation of liquid bridges with volume constraint the surface energies associated with the stable solutions also increases continuously when the contact angle is increased.

2. The unstable solutions appear when the contact angle is increased and it converges with the stable solution at a critical contact angle.

3. As the stable and unstable solutions reside on the same side of the critical point the bifurcation can be termed as subcritical. The stability margins can be calculated by taking the difference in surface energies associated with stable and unstable solutions for a certain contact angle.

4. The critical points of the contact angle appear to be independent of the volume of the liquid and lies in the range of $144^\circ - 148^\circ$. This could be an advantage for contact angle manipulation when the contact angles can work in that range.

5. When the contact angle is constricted to an angle below the critical point the release mechanism would depend on the stability margin. The volume of the liquid would play an important role here as the stability margin increases with increasing volume. If the contact angle of $70^\circ$ need to be used as the pickup angle and $120^\circ$ as the release angle then a choice of liquid volume should be such that the stability margin at $70^\circ$ is greater than energy associated with natural disturbance and stability margin for $120^\circ$ is less than that energy, or the energy is small enough to overcome by very small perturbations applied to the system.
6.5 Conclusion

We have studied the stability analysis of an axisymmetric liquid bridge between a spherical surface and a flat surface in this chapter. The classical stability analysis was pursued following the spectral stability criterion and the critical stability graph was plotted. Stable and unstable solutions corresponding to change in gap was satisfactorily explained but the solutions corresponding to change in contact angle remained unexplainable in the light of critical stability boundary. Another approach based on observation of plot of preferred coordinates was inconclusive for the case of contact angle manipulation as no turning point in PV diagram was observed. A number of new parameters have been defined which gave an insight into problem but a concrete guidance on making a judgment on stability of the liquid bridge could not be offered. Finally stability margin analysis proved to be useful in explaining the stability and instability of the liquid bridge depending on the free surface energy. The amount of energy required to move a stable solution to unstable solution was obtained for different liquid bridge volumes. This study gave the quantitative measurement of disturbance or perturbation energy required to break the liquid bridge which helped in proposing another form of release mechanism.
CHAPTER VII
SUMMARY AND CONCLUSION

7.1 Summary & Conclusions

A liquid bridge based microgripper has been designed in this work in order to contribute to the study of microassembly research. From the initial study it was found that adhesive forces poses a problem to conventional 2 point or 3 point contact based tweezer like microgrippers. A comparative force analysis revealed that the most dominant of these adhesive forces is capillary and surface tension forces. The idea of using these adhesive forces in favor of gripping the objects resulted in the idea of using a liquid bridge based microgripper.

To prove that this type of microgripper is indeed capable of picking up an object a study on the forces was needed. The capillary and surface tension force was found to be dependent on the shape of the liquid bridge, which in turn depends on a number of parameters, like size of the object, liquid volume, contact angle of liquid with two surfaces, gap between the object and the gripper surface. So as a first step, a framework was created that can give the shape of the liquid bridge when any of the design parameters is changed. The governing equations were solved using the boundary conditions and volume constraint by a triple iterative scheme implemented in Matlab. The numerical solution was matched with the catenoid profile which corresponds to
analytical solution of the meniscus profile for zero pressure difference. Simulation results for change in gap showed that the maximum lifting forces are 2-5 times of the weight of the object depending on the volume of liquid used, which proved the concept that liquid bridge based microgrippers can pickup an object. The important observation here was the effect of gap on the lifting forces. As the gap is increased two solutions were obtained by solving the nonlinear Young-Laplace equation, the solution corresponding to lower surface energy was termed as stable solution and the solution corresponding to higher surface energy is termed as unstable solution. It was found that these two solutions converge to a critical gap, after which the numerical simulation does not provide any solution. Previous experimental studies reported this point as a rupture point, where the liquid bridge breaks. The force diagram revealed that the lifting forces decreases from its maximum value when the gap is increased. Now when the gripper surface with the liquid droplet comes in contact with the object and the liquid bridge is formed, the gap between the object and the surface was assumed to be the height of the pendant droplet. This assumption provides us with the initial gap for the pickup step and later this gap has been used for contact angle manipulation for release. For contact angle manipulation a parametric study was conducted where the contact angle of the liquid and the gripper surface has been changed. When this contact angle is increased it was found that the lifting forces decrease monotonically. The contact angle of the liquid with the spherical surface of the object is assumed to be constant at 20°. The forces decreased below the weight of the object for a range of liquid volume. Also the appearance of unstable solutions was observed and a critical contact angle found, which was considered to be
similar to critical gap and this point was logically concluded be the rupture point. So it was found that by contact angle manipulation when the contact angle reaches a critical point (generally a contact angle in the range of $145^\circ - 150^\circ$), the liquid bridge would cease to exist. From this analysis two types of release mechanism can be proposed.

- If the gripper surface is coated with a Teflon coating which restricts the working range of contact angle to $120^\circ - 70^\circ$, then a dimensionless liquid volume of the range $0.01 - 1$ is sufficient for release of the object.

- If the gripper surface is coated with a super hydrophobic material with contact angle in the range $150^\circ$, the contact angle manipulation would break the liquid bridge at a rupture point when the contact angle reaches the critical contact angle.

Once it was understood that contact angle manipulation is indeed helpful in release mechanism the next step was to find a way to control the contact angle. Electrowetting was found to be a convenient technique for changing the contact angle and very recently a number of applications of liquid droplet manipulation were found to be reported in the MEMS community. Further study on this technique revealed the limitations. The major limitation was found to be contact angle saturation. The saturation effect limited the contact angle pickup to be in the range of $70^\circ$. Another limitation was found to be the placement of the ground wire, as two electrodes were needed for electrowetting. The solution was found in a single plate electrowetting design where the ground wire was placed inside the dielectric layer. The single plate electrowetting design was found to be perfect for gripper surface design where the liquid hangs from the gripper surface as a pendant droplet and later the object hangs from the gripper surface by
the liquid bridge. The physics behind electrowetting was studies and from the different saturation models it was understood that the actual cause of saturation effect is still unknown. The saturation effect cannot be described by a single theory and it might be due to combination of multiple theories depending on the experimental setup used.

To further validate the numerical solutions obtained by the solution of Young-Laplace equation, a commercial software CFD ACE+ was used. The simulation results obtained from CFD ACE+ matched exactly with the results obtained from numerical solution for smaller contact angles (i.e. 60°), but diverged slightly for higher contact angles (i.e. 120°). The divergence can be attributed to the viscous effect taken into consideration in the CFD Analysis. The CFD ACE+ software results were validated against an image of a droplet of volume 10μl and the results matched satisfactorily. The transient analysis if the CFD ACE+ solutions further gave information on the timescale of the angle transformations and it was observed that when the contact angle was increased to 140°, the liquid bridge ruptures at the interface and breaks. These two simulation studies proved the fact that contact angle manipulation can be effective in breaking the liquid bridge. But the contact angle at which the liquid bridge breaks or ruptures was found to be higher than the contact angle of Teflon. So of the gripper surface is coated with Teflon, with contact angle of 120°, the liquid bridge will not break. To understand the stability of the liquid bridge at this position further stability study was conducted.

At first a spectral stability criterion was formulated, where the sign of the eigenvalues of the perturbation equation corresponds to stability and instability of the
system. The critical stability boundaries for the systems were mapped on a nondimensional space and the stable unstable solution for gap change and angle change were plotted. Though the plot of the gap change can be explained in the light of critical stability, the plot of angle change could not be explained properly. A second approach for stability study was found to be by observing the change in direction of the preferred diagrams where the preferred coordinates corresponds to the pressure and volume of the system. Plots of solutions corresponds to the gap change again can be explained, but plot of solutions corresponding to angle change was remained unexplained. Finally the stability margin approach was used to explain the state of stability and instability. The stability margin was found to reduce when the contact angle is increased or when the volume is decreased. Stable state with less amount of stability margin was found to be vulnerable to slight disturbances, slight perturbation can change a stable state into an unstable state if the energy imparted on the systems exceeds the stability margin. This explanation of stability opened up the option of another release mechanism by slight disturbances.

Finally it can be concluded that the liquid bridge based microgripper is capable to pick up an object and release of the object is possible for through contact angle manipulation. Different mechanism can be used for the release as discussed earlier. Simulation results have proved the hypothesis of the manipulation scheme.
7.2 Future Study

This study deals with the theoretical aspects of the microgripper design so future direction for this research would obviously be the experimental validation. A prototype gripper needs to be made and pickup and release mechanism justified through rigorous experiments. Further study is needed to understand what the initial gap should be when the liquid bridge is formed at the time of pick up of the object. When the liquid volume is increased or the angle is increased, understanding the effect on the gap might change would be an important extension of this study. An effective liquid dispensing method needs to be designed which would be capable of delivering precise liquid volume on the surface of the gripper with picolitre precision. The effect of charge distribution and electric field on the liquid or the objects needs further investigation.
REFERENCES


Chapter 3


33. A. W. Adamson, “Physical Chemistry of Surfaces”, Published by Inter Science Publisher, 1963, pp. 5.


Chapter 4


Chapter 5


Chapter 6


194
82. Laplace, R. S., 1806, ”Traite de mechanique Celeste”, vol 10. Paris(coureier)


APPENDIX A:

DIFFERENTIAL EQUATION FORM OF LAPLACE EQUATION

The Laplace Equation can be expressed in differential equation form in terms of the meniscus geometry. Figure A.1 shows the meniscus geometry of a liquid bridge formed between a sphere of radius R and a plane placed at a distance D away from the sphere.

Proof

If the meniscus is axis-symmetric then the mean curvature of then follows the Laplace equation

\[
\frac{\Delta p}{\gamma} = 2H = \frac{1}{R_1} + \frac{1}{R_2} = \frac{\frac{d^2 z}{dr^2}}{1 + \left(\frac{dz}{dr}\right)^2} + \frac{\frac{dz}{dr}}{\sqrt{1 + \left(\frac{dz}{dr}\right)^2}}
\]

A.1

Where \( \Delta p \) is the pressure difference across the meniscus and \( H \) is the mean curvature of the meniscus, \( R_1 \) and \( R_2 \) are the principle radii of curvature.
Figure A.1 Meniscus Profile of liquid bridge formed between a sphere and a plane

Using non-dimensional variables \( y \) and \( x \) we can write

\[ y = \frac{z}{R} \quad \text{and} \quad x = \frac{r}{R} \]

Substituting in Equation A.2 in Equation A.1 we can get

\[
\frac{dy}{dx} = \frac{d}{d\left(\frac{r}{R}\right)} \left(\frac{z}{R}\right) = \frac{dz}{dr} \quad \frac{d^2y}{dx^2} = \frac{d}{d\left(\frac{r}{R}\right)} \frac{dz}{dr} = R \frac{d^2z}{dr^2}
\]

A.2
\[ 2H = \frac{1}{R} \frac{d^2 y}{dx^2} + \frac{dy}{dx} \left[ 1 + \left( \frac{dy}{dx} \right)^2 \right]^{\frac{1}{2}} \]

From geometry it is clear that

\[ \frac{dy}{dx} = \frac{dz}{dr} = \tan \varepsilon \quad \text{so} \quad 1 + \left( \frac{dy}{dx} \right)^2 = \sec^2 \varepsilon \quad \text{and} \quad \frac{d^2 y}{dx^2} = \sec^2 \varepsilon \frac{d \varepsilon}{dx} \quad \text{A.4} \]

Substituting Equation A.4 into Equation A.3 we get

\[ 2H = \frac{1}{R} \frac{\sec^2 \varepsilon}{\sec^3 \varepsilon} \frac{d \varepsilon}{dx} + \tan \varepsilon \frac{Rx \sec \varepsilon}{Rx \sec \varepsilon} = \frac{1}{R} \cos \varepsilon \frac{d \varepsilon}{dx} + \sin \varepsilon \frac{\sec \varepsilon}{Rx} \quad \text{A.5} \]

Substituting \( u = -\sin \varepsilon \) and \( \frac{du}{dx} = -\cos \varepsilon \frac{d \varepsilon}{dx} \) in Equation A.5 we get

\[ 2HR = -\frac{du}{dx} - \frac{u}{x} \quad \text{A.6} \]
APPENDIX B:

FORMULATION OF PARAMETRIC DIFFERENTIAL EQUATION

Aveyard [37] used a parametric form of the Laplace equation. It is not the same as
the above Equation A.6 as the parameters are not normalized in this case. The equation is
transformed to the form as shown below by using parametric method

\[
\frac{dr}{ds} = \cos \varepsilon \ ; \quad \frac{dz}{ds} = \sin \varepsilon \ ; \quad \frac{d\varepsilon}{ds} = \frac{\Delta p}{\gamma} - \frac{\sin \varepsilon}{r} \tag{B.1}
\]

where \( s \) is the length of the capillary bridge curve.

Proof

Let us take the meniscus profile as a function \( r = r(z) \) then \( r'(z) = \cot \varepsilon \) where \( \varepsilon \) is the
angle made by the normal to the meniscus with vertical axis.

Then from Figure A.1 it can be seen that \( \frac{dz}{dr} = \tan \varepsilon \)

\[
2H = \frac{d^2z}{dr^2} + \frac{dz}{dr} \left[ 1 + \left( \frac{dz}{dr} \right)^2 \right]^{\frac{3}{2}} = \frac{d^2\varepsilon}{dr^2} \left[ 1 + (\tan \varepsilon)^2 \right]^{\frac{3}{2}} + \frac{\tan \varepsilon}{r \left[ 1 + (\tan \varepsilon)^2 \right]^{\frac{3}{2}}} \tag{B.2}
\]

\[
2H = \frac{\sec^2 \varepsilon}{\sec^3 \varepsilon} \frac{d\varepsilon}{dr} + \tan \varepsilon = \cos \varepsilon \frac{d\varepsilon}{dr} + \frac{\sin \varepsilon}{r} = \frac{1}{r} \frac{d}{dr} (r \sin \varepsilon) \tag{B.3}
\]

From Laplace equation we know \( \Delta p = 2H \gamma \) so we can directly write
\[
\frac{\Delta p}{\gamma} = \cos \varepsilon \frac{d\varepsilon}{dr} + \frac{\sin \varepsilon}{r} = \frac{dr}{ds} \frac{d\varepsilon}{dr} + \frac{\sin \varepsilon}{r} \quad \text{where} \quad \frac{dr}{ds} = \cos \varepsilon \text{ and } \frac{dz}{ds} = \sin \varepsilon \quad \text{B.4}
\]

The parametric equations take the form as

\[
\frac{dr}{ds} = \cos \varepsilon \quad ; \quad \frac{dz}{ds} = \sin \varepsilon \quad ; \quad \frac{d\varepsilon}{ds} = \frac{\Delta p}{\gamma} - \frac{\sin \varepsilon}{r} \quad \text{B.5}
\]
APPENDIX C:

NOTE ON LEGENDRE CONDITION

A necessary condition for the solution of the simplest problem in variational calculus, proposed by A.M. Legendre in 1786: For the curve $y_0(x)$ to provide a minimum of the functional

$$J = \int F(x, y, y')dx, \quad y(x_1) = y_1, \quad y(x_2) = y_2$$  \hspace{1cm} \text{C.1}$$

it is necessary that at all points of $y_0(x)$ the second derivative of the integrand with respect to $y'$ should be non-negative:

$$F_{y'y'}(x, y_0(x), y_0'(x)) \geq 0, \quad x_1 \leq x \leq x_2$$  \hspace{1cm} \text{C.2}$$

If $y$ is an $n$-dimensional vector with coordinates $y_1 \ldots y_n$, then the Legendre condition requires that the quadratic form

$$\sum_{i=1}^{n} \sum_{j=1}^{n} F_{y_i'y_j'}(x, y_0(x), y_0'(x))\eta_i\eta_j \geq 0, \quad x \in [x_1, x_2], \quad \eta = (\eta_1, \ldots, \eta_n) \in \mathbb{R}^n$$  \hspace{1cm} \text{C.3}$$
should be non-negative. For the case of a maximum of the functional the sign of the inequality in the Legendre condition is reversed. For variational problems on a conditional extremum the analogue of the Legendre condition is the Clebsch condition.

The Legendre condition, like the Euler equation, is a necessary condition for a weak extremum. If the Legendre condition is violated, the second variation of the functional does not preserve its sign and the curve $y_0(x)$ does not provide an extremum of the functional.

If in the Legendre condition the sign of non-strict inequality is replaced by the sign of strict inequality, then the condition is called the strong Legendre condition. The strong Legendre condition, in contrast to the Legendre condition, is not necessary. The strong Legendre condition is involved in the formulation of sufficient conditions for an extremum. An extremal on which the strong Legendre condition is satisfied is called a non-singular extremal. Such an extremal is twice continuously differentiable, and the Euler equation for it can be represented as an ordinary differential equation of the second order, solved for the highest derivative. If the strong Jacobi condition is satisfied on a non-singular extremal, then one can construct a field of extremals surrounding the given extremal, which is the first step in the investigation of sufficient conditions for an extremum. Ref: [http://eom.springer.de/L/l058010.htm](http://eom.springer.de/L/l058010.htm)
The first equation for the spectral stability criterion corresponding to the axisymmetric perturbation is

\[- \varphi_0'' - \frac{r'}{r} \varphi_0' + a(s) \varphi_0 + \mu = \lambda \varphi_0 \quad 0 \leq s \leq s_1\]  \hspace{1cm} D.1

with boundary condition

\[\left( - \varphi_0' + \chi \varphi_0 \right)_{s=0} = 0, \quad \left( \varphi_0' + \chi \varphi_0 \right)_{s=s_1} = 0, \quad \int_0^{s_1} r \varphi_0(s) ds = 0\]  \hspace{1cm} D.2

Equation D.1 can be written as

\[- \varphi_0'' - \frac{r'}{r} \varphi_0' + a(s) + \frac{\mu}{\varphi_0} = \lambda \quad 0 \leq s \leq s_1\]  \hspace{1cm} D.3

at \(s = s_1\) the boundary condition can be written as \(\chi\big|_{s=s_1} = -\frac{\varphi_0'}{\varphi_0}\) and substituting it in gives

\[- \varphi_0'' + \frac{r'}{r} \chi\big|_{s=s_1} + a(s) + \frac{\mu}{\varphi_0} = \lambda\]
From the form of this equation it can be concluded that if functions $a(.)$ and $\chi$ assumes higher values $\lambda^*$ will increase. $\lambda^*$ is defined as the minimum eigenvalue of the two set of solutions $\lambda^* = \min(\lambda_{01}, \lambda_{11})$ where $\lambda_{01}, \lambda_{11}$ are solutions of (5.16) and (5.18) with boundary conditions (5.17) and (5.19). So for a fixed $\Gamma$ and a function $a(.)$ it can be verified that $\lim_{x_i \to -\infty} \lambda^* = -\infty$ and if $a(.)$ has a upper bound then $\lim_{x_i \to +\infty} \lambda^* = \nu^*$ where $\nu^*$ is the eigen value of the analogous problem with boundary condition $N|_{\nu} = 0$. In particular it can be concluded that the value of $\lambda^*$ has an upper bound $\lambda^* \leq \nu^*$ where $\nu^*$ is the lowest eigenvalue of a similar problem with boundary condition $N|_{\nu} = 0$ and $\nu^*$ does not depend on the value of $\chi$. 
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