

AN EXPERIMENTAL STUDY ON THE STABILITY OF THE INTERFACE
BETWEEN IMMISCIBLE LIQUIDS IN A MICRO CHANNEL SUBJECT TO AN
ELECTRIC FIELD

by

Pınar Eribol

B.S., Chemical Engineering, Yeditepe University, 2012

Submitted to the Institute for Graduate Studies in
Science and Engineering in partial fulfillment of
the requirements for the degree of
Master of Science

Graduate Program in Chemical Engineering

Boğaziçi University

2014

ACKNOWLEDGEMENTS

First and foremost, I would like to express my sincere gratitude to my supervisor Assoc. Prof. A. Kerem Uğuz for his guidance, patience, motivation, and enthusiasm throughout my research.

Besides, I am deeply grateful to Prof. Ramazan Yıldırım for his important help, support and guidance. Prof. Yıldırım has always welcomed my questions with a smile and helped me during my research.

My sincere thanks go to Assoc. Prof. Şenol Mutlu and his research group for their valuable time and helping us to improve this research. I also want to thank Assist. Prof. Oktay Demircan for his help and letting us work in his laboratory.

I would like to acknowledge the guidance and support of all of my professors. I also want to thank Bilgi Dedeoğlu and Yakup Bal for their technical assistance, and Melike Gürbüz, Belgin Balkan for their kindness and help.

I am very grateful to my friends, Çiğdem Ekmen and Hazal Bal for not leaving me alone during this long research. They have made our time in Boğaziçi more enjoyable. Without motivating each other, it would be impossible to overcome this mental process easily. I am also very thankful to my friends from Yeditepe, they have never stopped supporting and trusting me.

My last thanks are to the people who helped me most to survive this process mentally. Eshan Newaz, thanks for not letting me give up and listening to me all the time. I also owe my thanks to my grandparents for always believing me. I would also like to thank my parents for supporting me both spiritually and materially through all my life. They have always encouraged me patiently and tolerantly at all of the good and bad times. Finally, this thesis is dedicated to my brother, Okan Eribol whom I love the most.

The financial support for this thesis was provided by TÜBİTAK Project 109M557 and Boğaziçi University Research Fund Project 8085.

ABSTRACT

AN EXPERIMENTAL STUDY ON THE STABILITY OF THE INTERFACE BETWEEN IMMISCIBLE LIQUIDS IN A MICRO CHANNEL SUBJECT TO AN ELECTRIC FIELD

Micro channels offer many advantages for both industrial and laboratory applications. They provide the use of small amounts of sample, reduced cost and faster analysis. The flow regime is laminar in micro channels. Electrohydrodynamic (EHD) instability can be used in micro channels to obtain an efficient mixing for miscible liquids and to generate mono dispersed droplets for immiscible liquids. In this study, the purpose is to experimentally destabilize the flat interface between two immiscible and Newtonian liquids flowing in a rectangular micro channel in the presence of an electric field. The channels are of depth 0.2 mm and width 1.0 and 1.5 mm. The liquids are pumped into the micro channel with a syringe pump and a DC electric field is applied either parallel or normal to the flat interface formed between these immiscible liquids. The liquid pairs used in the experiments are a combination of ethylene glycol, silicone oils with different viscosities, castor oil, and olive oil. Electrohydrodynamic instability is investigated for several parameters including the ratios of viscosities and flow rates, the width of the micro channel and the direction of the electric field. The results of the experiments show that an increase in viscosity and flow rate ratios of silicone oil to ethylene glycol has a stabilizing effect in the presence of a normal electric field. In addition, it is found that only the ratio of the flow rates has an effect on the stability of the interface, not the individual flow rates of the liquids. Furthermore, an increase in the width of the channel results in an increase in the critical potential difference of the system. The range of voltages needed to destabilize the interface in the performed experiments are in between 94 and 1090 volts. Finally, it is shown that the parallel electric field does not have a destabilizing effect on the liquid pairs used in the study.

ÖZET

BİR MİKRO KANALDA AKMAKTA OLAN KARIŞMAYAN SIVILAR ARASINDAKİ ARAYÜZEYİN ELEKTRİK ALAN ETKİSİNDEKİ KARARLILIĞININ DENEYSEL OLARAK İNCELENMESİ

Mikro kanallar endüstriyel ve laboratuvar uygulamalarında pek çok avantaj sunmaktadır. Bu kanallar, az miktarda numune kullanımı, düşük maliyetli ve daha hızlı analiz imkanı sağlamaktadır. Mikro kanallardaki akış kanal boyutundan dolayı laminerdir Elektrohidrodinamik (EHD) kararsızlık, mikro kanallarda karışabilen sıvılar için etkili bir karışım elde etmek ve karışmayan sıvılar için mikro damlacıklar oluşturmak amacı ile kullanılabilir. Bu çalışmada, amaç dikdörtgen bir mikro kanalda akmakta olan iki karışmayan ve Newtonyen tipi sıvının arasında bulunan düz arayüzeyin elektrik alan etkisinde kararsız hale getirilmesidir. Mikro kanalların derinliği 0.2 mm, genişlikleri 1.0 ve 1.5 mm'dir. Sıvılar, bir şırınga pompası ile mikro kanal içine pompalanmakta ve bu sıvılar arasında oluşan düz arayüzeye DC elektrik alanı paralel veya dik olarak uygulanmaktadır. Deneylerde kullanılan sıvı çiftleri etilen glikol, farklı viskozitelerde silikon yağları, hint yağı ve zeytinyağından oluşmaktadır. Elektrohidrodinamik kararsızlık viskoziteler ve akış hızlarının oranları, mikro kanalın genişliği ve elektrik alanın yönü de dâhil olmak üzere pek çok değişken bakımından araştırılmıştır. Deneylerin sonuçları dik uygulanan elektrik alanının varlığında etilen glikolün viskozite ve akış hızının silikon yağının viskozite ve akış hızına oranlarındaki bir artışın, karışmayan sıvılar arasında oluşan arayüzeyin üzerinde kararlı hale getirici bir etkiye sahip olduğunu göstermektedir. Ek olarak, sıvıların kendi akış hızlarının değil, akış hızlarının birbirine olan oranlarının arayüzeyin kararlılığı üzerinde bir etkiye sahip olduğu bulunmuştur. Ayrıca, mikro kanalın genişliğindeki değişimin arayüzeyin kararlılığına olan etkisinin araştırıldığı çalışmaların sonuçları göstermektedir ki kanalın genişliğindeki artış, sistemlerin kritik potansiyel farkında artışa yol açmaktadır. Deneylerde arayüzü kararsız hale getirmek için gerekli potansiyel fark aralığı 94 ve 1090 volt arasında bulunmaktadır. Son olarak, arayüzeye paralel olarak uygulanan elektrik alanının, deneylerde kullanılan sıvı çiftleri üzerinde kararsız hal yaratacak bir etkiye sahip olmadığı gösterilmiştir.

TABLE OF CONTENTS

ACKNOWLEDGEMENTS	iv
ABSTRACT	vi
ÖZET	vii
LIST OF FIGURES	x
LIST OF TABLES	xii
LIST OF ACRONYMS/ABBREVIATIONS	xiii
LIST OF SYMBOLS	xiiiv
1. INTRODUCTION	1
2. LITERATURE SURVEY	4
2.1. Applications of Droplet Microfluidics	4
2.2. Droplet Formation in Micro Channels	6
2.2.1. Passive Methods for Droplet Formation in Micro Channels	6
2.2.2.1. Droplet Formation in Co-flowing streams	6
2.2.2.2. Droplet Formation in a T-shaped Junction	7
2.2.2.3. Droplet Formation in Flow Focusing Streams	9
2.2.3. Active Methods for Droplet Formation in Micro Channels	10
2.2.3.1. Thermal Actuation	10
2.2.3.2. Pressure Actuation	11
2.2.3.3. Electro-Magnetic Field Application	12
2.2.3.4. Electric Field Application	12
2.3. Electrohydrodynamic Instabilities	14
2.3.1. The Leaky Dielectric Model	16
2.3.2. Direction of the Applied Electric Field	17
2.3.3. Direct Current or Alternating Current	18

3. EXPERIMENTAL WORK	20
3.1. Chemicals	20
3.2. Experimental Set-up	22
3.3. Procedure of the Experiment	25
4. RESULTS AND DISCUSSION	30
4.1. Evolution of the Electrohydrodynamic Instability	30
4.2. The Effect of the Viscosity Ratio	32
4.3. The Effect of the Flow Rate Ratio	35
4.3.1. The Effect of the Changing Flow Rate Ratio.	35
4.3.2. The Effect of Constant Flow Rate Ratio	38
4.4. The Effect of the Channel Width	40
4.5. The Effect of the Direction of the Electric Field	40
5. CONCLUSIONS AND RECOMMENDATIONS	42
5.1. Conclusions	42
5.2. Recommendations	43
REFERENCES	45

LIST OF FIGURES

Figure 2.1.	The droplet formation in a channel with a co-flowing device (Adapted from Nunes <i>et al.</i> , 2013)	7
Figure 2.2.	Droplet formation in a channel with a T-junction (Adapted from Nunes <i>et al.</i> , 2013)	8
Figure 2.3.	Droplet formation with a flow focusing device (Adapted from Nunes <i>et al.</i> , 2013)	10
Figure 3.1.	Sketch of the experimental setup	22
Figure 3.2.	Experimental set-up	23
Figure 3.3.	Channel geometry (a) normal electric field with a width of 1.0 mm (b) normal electric field with a width of 1.5 mm (c) parallel electric field with a width of 1.0 mm	24
Figure 3.4.	Micro channel with parallel electric field application	24
Figure 3.5.	Micro channel with a width of 1.0 mm normal electric field application	25
Figure 3.6.	Micro channel with a width of 1.5 mm normal electric field application	25
Figure 3.7.	The flat interface between silicone oil, 50 cSt and ethylene glycol; both with a flow rate of 40 μ L/min	26
Figure 3.8.	The critical voltage detected for the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min at 725 V	27
Figure 3.9.	The increased instability at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min at 725 V	27
Figure 3.10.	The rupture at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min	28

Figure 3.11.	Complete rupture at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min	28
Figure 3.12.	The interface of the silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min the voltage is decreased below its critical value	29
Figure 4.1.	The instability occurred at the interface between ethylene glycol and silicone oil, 50 cSt at a critical potential difference of 880 V with respect to time in the presence of a normal electric field	31
Figure 4.2.	The effect of the viscosity ratio on the voltage in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b)	33
Figure 4.3.	The effect of the viscosity ratio on the electric number, E_b in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b)	34
Figure 4.4.	Effect of the flow rate ratio on the potential difference in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b)	36
Figure 4.5.	The effect of the flow rate ratio on the electric number in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b)	37
Figure 4.6.	The effect of the same flow rate ratio of unity for silicone oil-ethylene glycol pairs on the voltage (a) and electric number, E_b (b) in the presence of a normal electric field	39
Figure 4.7.	The effect of the width of the channel for silicone oil-ethylene glycol pairs on the voltage (a) and electric number, E_b (b) in the presence of a normal electric field	41

LIST OF TABLES

Table 3.1. The chemicals used in the experiments	17
Table 3.2. Physical properties of the chemicals used in the experiments	18

LIST OF ACRONYMS/ABBREVIATIONS

AC	Alternative current
DC	Direct current
EHD	Electrohydrodynamic
ESX	Ethylene glycol – Silicone oil with viscosity of X
PMMA	Poly(methyl methacrylate)
E_b	Dimensionless electric number
H	Width of the micro channel
$H^{(i)}$	Height of the i^{th} phase
H_r	Height ratio
Pé	Péclet number
$Q^{(i)}$	Flow rate of the i^{th} phase
Q_r	Flow rate ratio
Re	Reynolds number
T	Time
U	Characteristic velocity
V_b	Applied voltage
V	Voltage

LIST OF SYMBOLS

Γ	Surface tension
E	Electrical permittivity
ϵ_0	Vacuum Permittivity
M	Dynamic Viscosity
μ_r	Viscosity ratio
N	Kinematic viscosity
P	Density
Σ	Electrical Conductivity

1. INTRODUCTION

Microfluidics is the science and technology that deals with devices and methods that can control fluids in micro channels with a typical length scale of about hundred nanometers to several micrometers. Microfluidics technologies have first been applied to analyze and carry out separations and detection technology, which allow the usage of small amounts of sample, reduced cost and faster analysis under better conditions such as high resolution and sensitivity (Whitesides *et al.*, 2006).

The flow in the micro channels is usually laminar due to low Reynolds number ($Re < 100$) and the Péclet number is large ($Pé > 100$) (Stroock *et al.*, 2002). In such systems the turbulence does not occur and for the miscible liquids the mixing is limited by the molecular diffusion since the process is extremely slow (Schwesinger *et al.*, 1996, Stone *et al.*, 2004 and Fu *et al.*, 2004). Conventional mixing methods that are used to handle larger volumes are often not practical in micro scale. For that reason, micro channels require different techniques than the conventional ones. In micro channels the mixing is enhanced by two types of methods, one is the passive methods that include complex geometries (Hosokawa *et al.*, 1996; Liu *et al.*, 2000), pulse flows or micro mixers (Yang *et al.*, 2007) and do not require an energy input to achieve the mixing. The second type of mixing technique is via active methods (Moroney *et al.*, 1991; Evans *et al.*, 1997; Evensen *et al.*, 1998), which require external forces such as magnetic or electric forces to enhance the mixing in a micro sized device. An electric field can be coupled with magnetic field and they can induce the mixing in a micro channel using the electrodes located on the walls of a micro channel (West *et al.*, 2000). In this work, the mixing of the miscible liquids will not be discussed. For a detailed study of mixing of miscible fluids, the works of Liu *et al.* (2000) and Hessel *et al.* (2004) are suggested.

Another research area of the micro channel studies include droplet formation. Micro droplet can be generated from immiscible liquids and it can be used in many fields such as in chemical industry (Hatakeyema *et al.*, 2006) or in biology (deMello, 2001; Vogelstein and Kinzler, 1999; Zheng *et al.*, 2004; Song and Ismagilov, 2003). To produce a droplet, the system should include at least two immiscible fluids where their flow is controlled by

volume, pressure, or vacuum. First, an interface is created between immiscible fluids, then this interface is disturbed and a droplet occurs when one of the immiscible fluids is dispersed into the continuous phase. Recently, it has been developed many methods to form droplets in micro channels. As the methods used in the mixing of the miscible liquids, the methods used to generate droplets are also categorized as passive and active methods. In passive methods, the surface is induced by modified channel geometries like co-flowing streams (Umbanhowar *et al.*, 2000), T-shaped junctions (Thorsen *et al.*, 2001) and flow focusing (Anna *et al.*, 2003). Active methods can be used to create micro droplets with uniform size distribution easily. However, these methods need an external actuator as pressure (Willaime *et al.*, 2006), magnetic field (Tan *et al.*, 2010), temperature (Nguyen *et al.* 2007; Murshed *et al.* 2008a) or electric field. Ozen *et al.* (2006a) created mono dispersed droplets from immiscible liquids in a micro channel by applying a DC electric field normal to the interface between these liquids. When an electric field is applied to the interface, electrohydrodynamic instability occurs. The interface starts deflecting at some voltage, called the critical voltage, and then the interface flaps one of the walls of the micro channel, and forms a droplet.

The electrohydrodynamic instability has been used in various studies (Baygents and Baldessari, 1998; Craster and Matar, 2005; Thaokar and Kumaran, 2005). The discontinuities in both the electrical conductivity and the permittivity cause the electrohydrodynamic instability for either leaky dielectric or perfect dielectric liquids (Taylor and Melcher, 1969; Saville, 1997). Linear stability analysis of the interface between two immiscible fluids in the presence of an electric field is examined in several works for perfect and leaky dielectric fluids (Ozen *et al.*, 2006; Uguz and Aubry, 2008; Shankar and Sharma, 2004). The electric field is applied either parallel (Patankar, 2011) or normal (Li *et al.* 2011) to the interface and the results of both cases are compared (Uguz *et al.*, 2008; Hsu and Patankar, 2014).

In this study, the stability of the interface between two immiscible and Newtonian liquids is investigated experimentally in the presence of an electric field applied either normal or parallel to the flat interface between the immiscible liquids. The critical voltage at which the interface starts deflecting is determined as a function of viscosity and height ratios of liquids, the channel width and the direction of the electric field. This thesis starts

with the literature survey on the formation techniques and applications of micro droplets and continues with electrohydrodynamics. In the experimental section, the experimental procedure is explained in detail. In the following section, the experimental results are introduced and discussed. Finally, in the last section, conclusions and recommendations are presented.

2. LITERATURE SURVEY

In the proceeding sections of this chapter, some of the various applications of the microdroplets will be briefly explained and different droplet formation methods in micro channels such as active and passive methods will be discussed. Since the effect of the electric field on the interface between two leaky dielectric liquids is studied in this thesis, the electrohydrodynamic instabilities and the leaky dielectric model will be reviewed. Finally, the effect of the direction of the electric field and DC/AC electric field are introduced.

2.1. Applications of Droplet Microfluidics

Nowadays droplet formation in micro channels is a promising research area since the micro droplets offer many advantages for both industrial and laboratory uses. It is possible to work in cells, molecules or reactors with very small volumes and each species or reactions can be isolated and controlled independently at desired conditions.

For chemical purposes, it is reasonable to use micro droplets since the chemical reagents can be expensive, dangerous to use at macro scale or limited in source. In biology, it is required to perform multiple reactions or detections in a short time and these requests can be achieved by using droplets. It is important to have a homogeneous environment for both chemical and biological reactions and the mixing in the droplets is the most effective way to achieve this. The droplets are used as micro reactors. For example, organic molecules (Hatakeyema *et al.*, 2006) or nanoparticles can be synthesized in micro droplet reactors. Of course, the yield of the product and the size distributions are enhanced due to high mixing and ability to adjust the reagent addition in the droplets. One of the most successfully and commercially applied biological methods is the emulsion polymerase chain reaction (PCR). Using the droplets prevents the interaction of polymerases and DNA templates with the channel walls. Polymerase can be inactivated by the interaction with the channel wall because of the contamination of the samples (de Mello, 2001). Monoclonal is a droplet that contains a single DNA molecule and a monoclonal can provide a chance to detect the medical problems such as pathogens and viruses by using only a small amount

of sample (Vogelstein and Kinzler, 1999). The droplets used to be polydisperse but now using the active droplet formation methods, it is possible to achieve a polydispersity of less than 1%. Thus, single cell or single molecule experiments can be done repeatedly at same conditions and large number of experimental data can be collected (Cai *et al.*, 2006).

Protein crystallization is used to study the structure of a protein by X-ray crystallography. When the protein solution becomes supersaturated, the proteins form crystals. However, protein samples are generally limited in sources. Due to this problem, the amount of protein samples can be easily be reduced to 4 nL to 10 μ L for protein crystallization experiments using nanoliter-sized droplets (Song *et al.* 2006; Zheng *et al.*, 2004). Working with small volume of samples does not indicate that the samples are dilute. When a single cell is stored in a droplet, the confinement of the cell causes into an increase in the density of the cell and the release of molecules. Thus, the increase in the cell density decreases the time needed to detect the molecules released by the cell (Song *et al.*, 2006).

Song and Ismagilov (2003) investigated the enzyme kinetics in droplets and the kinetics of the enzyme ribonuclease were measured in milliseconds. Also, the enzymatic activity of alkaline phosphatase and luciferase were studied in droplets (Roach *et al.*, 2005; Liao *et al.*, 2005). It is also possible to encapsulate a single or couple of cells into droplets and use the droplets as vessels (Kohler and Henkel, 2005). Furthermore, these droplets can be used to learn the network of the encapsulated cell (Breslauer *et al.*, 2006).

An important advantage of working with the droplets is the ability of preventing clogging of the channel from the precipitation. This was first studied by Poe *et al.* (2006). They studied the formation of indigo, amide and imine in the channels which results into precipitation. They were able to keep the precipitation in the dispersed phase and prevent the clogging of the channel from the precipitation. The mass transfer rate is enhanced in micro channels compared to macro scale devices due to the high surface to volume ratio and it is very important for heterogeneous reactions. The effect of the mass transfer was studied by Onal *et al.* (2005) on the catalytic hydrogenation of unsaturated aldehydes reaction.

2.2. Droplet Formation in Micro Channels

In droplet microfluidics the idea is that the production and the use of micro to nano size droplets from immiscible fluids in a controllable and reproducible manner. To produce a micro droplet, the system should include at least two immiscible fluids where their flow is controlled by pressure, volume, or vacuum. First, an interface is created between the immiscible fluids, then this interface is disturbed and the droplet formation occurs when one of the immiscible fluids (dispersed phase) is dispersed into the other immiscible fluid (continuous phase). Various methods are developed to form droplets in micro channels and these methods may be divided into two categories as passive and active methods.

2.2.1. Passive Methods for Droplet Formation in Micro Channels

In passive methods, the surface is induced by different geometries and the interface between the immiscible fluids is deformed by the flow field causes into a growth in the interfacial instabilities. No explicit external actuation and moving parts are used in the passive methods. The micro droplets have a size distribution with standard deviations of 1-3% for passive method applications and formed with a frequency of 2 to 5300 Hz (Gu *et al.*, 2011; Teh *et al.*, 2008). Passive methods can be categorized as breakup in co-flowing streams (Umbanhowar *et al.*, 2000), T-shaped junctions (Thorsen *et al.*, 2001) and flow focusing (Anna *et al.*, 2003).

2.2.2.1. Droplet Formation in Co-flowing streams. In the micro channels with co-flowing devices, the dispersed phase is driven into the continuous phase through an inner channel or a capillary with a small diameter and the dispersed phase is emerged from the capillary into the parallel flowing continuous phase. In the co-flowing streams, there are two separate breakup regimes i.e. jetting and dripping regimes. In the dripping regime, the droplet breaks up from an extended liquid jet, whereas in the dripping regime the droplet is generated at the tip of the capillary (Utada *et al.*, 2007). Figure 2.1. shows the droplet formation due to Rayleigh-Plateau instability in the dripping regime (Nunes *et al.*, 2013).

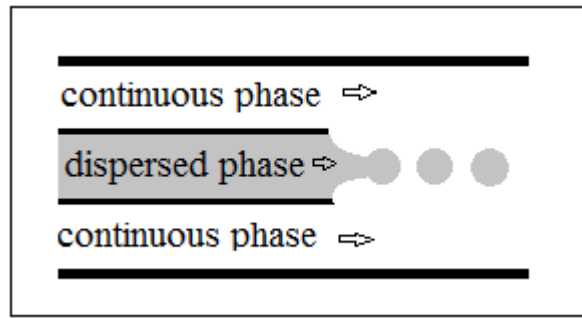


Figure 2.1. The droplet formation in a channel with a co-flowing device (Adapted from Nunes *et al.*, 2013).

Umbanhowar *et al.* (2000) experimentally studied the droplet formation from immiscible fluids in co-flowing streams. The droplet size is precisely achieved by changing the velocity of the continuous phase. Also the size of the droplets can be predicted by a model that includes parameters such as the velocity, the interfacial tension and the capillary diameter. In a study of Cramer *et al.* (2004), same parameters are studied and found that the interfacial tension is the only force making the droplet to have a spherical shape and prevents the formation of jets at the tip of the capillary. They concluded that the droplets with smaller size are formed for low interfacial tension and high flow rate of continuous phase and smaller flow rates of dispersed phase. It is observed that the effect of the viscosity of the dispersed phase is negligible for the size of the droplet.

2.2.2.2. Droplet Formation in a T-shaped Junction. A T-junction was first demonstrated by Thorsen *et al.* (2001) to produce monodisperse droplets in micro channels. In T-shaped junctions, the dispersed phase is perpendicularly introduced through a micro channel into the continuous phase flowing in the main micro channel (Thorsen *et al.*, 2012). Shear forces caused by the cross-flowing continuous phase and the pressure gradient results into a neck formation from the dispersed phase that finally thins and breaks up into droplets. For cross-flowing T-shaped junctions, three regimes are introduced as squeezing, dripping and jetting regimes (Garstecki *et al.*, 2006). Due to the physical restrictions of the micro devices, the squeezing regime is specific to the micro scale geometry at low capillary numbers. The dripping regime occurs at high capillary number when the shear and surface tension forces are balanced. Finally, the jetting regime is observed when the inertial forces

exceed the surface tension forces at high flow rates. (de Menech *et al.*, 2008; Utada *et al.*, 2007). Experimentally, droplets can be generated with a polydispersity of less than 1% in T-shaped junctions (Utada *et al.*, 2007). Furthermore, double emulsions are formed as water-in-oil-in-water or oil-in-water-in-oil using microfluidic devices having double T-junctions (Okushima *et al.*, 2004).

Garstecki *et al.* (2006) found that the length of droplet increases with decreasing of the continuous phase flow rate and it was possible to create droplets in micro scale when the capillary number is much less than unity. Also, if the width of the main micro channel is larger than its height and the width of the dispersed phase micro channel is at least half of the width of the main micro channel, the squeezing mechanism will be promoted. The droplet generation seen in Figure 2.2 is easy, regular and periodic for a single T-junction micro channel (Nunes *et al.*, 2013); although, it was observed that the droplet formation becomes less regular if multiple dispersed phase T-shaped channels are used (Barbier *et al.*, 2006; Garstecki *et al.*, 2005).

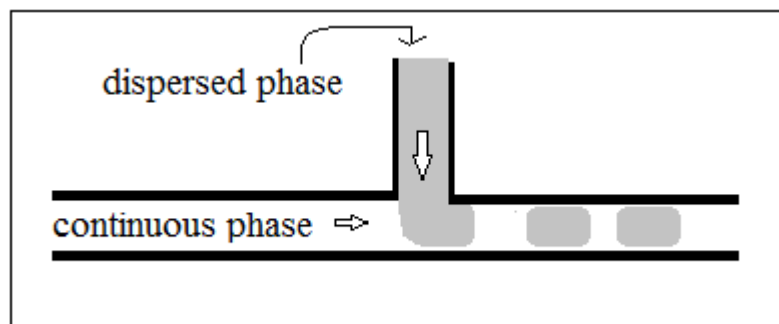


Figure 2.2. Droplet formation in a channel with a T-junction (Adapted from Nunes *et al.*, 2013).

In the study of Xu *et al.* (2005), a 45 μm square channel is connected to a 1 mm square main channel and the size of the generated droplets are measured within the range of 100-800 μm . In the same work, the effect of the continuous phase's velocity on the droplet size is investigated and observed that the droplet size decreases with increasing velocity of the continuous phase. Moreover, mono dispersed droplets can be generated

with a size range of 50-500 μm with a polydispersity of less than 2% (Xu *et al.*, 2006a; 2006b).

Experimental studies of the influence of the viscosity ratio show that the droplet size decreases when the viscosity of the continuous phase increases (Husny and Cooper-White, 2006; Xu *et al.*, 2006). On the other hand, the dispersed phase has an effect on the capillary number and as the viscosity ratio increases, the critical capillary number of the system increases, since the stability of the interface is enhanced by a higher internal viscosity (Tice *et al.*, 2004; Guillot and Colin, 2005).

2.2.2.3. Droplet Formation in Flow Focusing Streams. The flow focusing geometry was first used by Gañán-Calvo (1998) with a 3D axi-symmetrical configuration. Later, a planar version of this geometry was fabricated and easily inserted in a microfluidic device by Anna *et al.* (2003) using soft-lithography techniques. Four regimes are introduced for flow focusing streams as squeezing, dripping, jetting and thread formation (Kumacheva and Garstecki, 2011). Mono dispersed bubbles in a liquid can be prepared within a range of 10 to 1000 μm (Garstecki *et al.*, 2004). In Figure 2.3., the droplet formation is achieved by a flow focusing device.

In flow focusing devices, satellite droplets can be observed after the primary droplet formation. Tan and Lee (2005) reported a passive device to filter these satellite droplets forming in the flow focusing microfluidics device.

Garstecki *et al.* (2005) experimentally showed that the inertia of the flow dictates the stability of the interface. Romera and Abate (2012) recorded that the plugging-squeezing process causes the droplet formation for low to moderate capillary numbers. The size of the droplets depends on the viscosities of both the dispersed and the continuous liquids, the interfacial tension and the channel size (Hua *et al.*, 2007; Umbanhowar *et al.*, 2000).

Nie *et al.* (2008) investigated the effect of the viscosity on the droplet size and they found that the droplet size is considerably affected by the viscosity of the flow focusing streams. Although a simple scaling law cannot be proposed to predict the droplet size due

to complex flow field that is caused by the orifice resulting in increased number of geometrical aspect ratio (Baroud *et al.*, 2010).

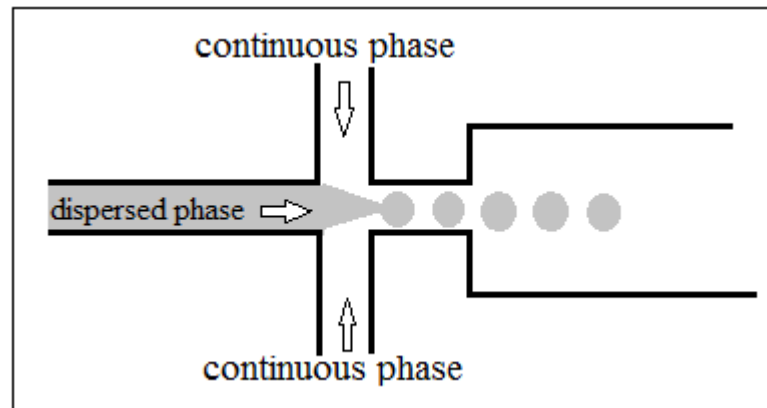


Figure 2.3. Droplet formation with a flow focusing device (Adapted from Nunes *et al.*, 2013).

Finite element simulation of droplet formation in microfluidics flow focusing device was investigated by Zhou *et al.* (2006) and their numerical results on the droplet size changing with the flow rate ratio matched with the experimental results. (Utada *et al.*, 2005).

2.2.3. Active Methods for Droplet Formation in Micro Channels

Droplets can also be generated by active breakup methods. An external force is applied to initiate the pinch off of the droplet that gives an extra manipulation for the droplet breakup frequency and the droplet size. Some of the external actuation methods for the droplet formation in microfluidics systems are thermal effects, pressure and piezoelectric actuations, magnetic and electric field applications.

2.2.3.1. Thermal Actuation. The droplet diameter is a function of flow rate, interfacial tension, and fluid viscosities where the interfacial tension and the fluid viscosity depend exponentially on the temperature. Using the temperature dependency of the viscosity and the interfacial tension, the droplet size was increased compared to its original value for

temperatures changing from 25 to 70°C. The exponential dependence was found to be valid for high temperature differences since the two different regimes occur for a temperature difference of 10 to 20 K (Nguyen *et al.*, 2007).

The effect of the temperature on droplet formation and size manipulation in a PDMS microfluidics T-junction is studied by Murshed *et al.* (2008a). Results show that the droplet size is directly proportional to temperature. In addition, the increase in the droplet size becomes larger for an increasing temperature at smaller micro channel depths. In another work of Murshed *et al.* (2008b), the droplet size and manipulation were experimentally investigated in a heat induced flow focusing micro device. Three different flow regimes were observed as Nguyen *et al.* (2007) did and these regimes were successfully controlled by a temperature change from 25°C to 42° C (Murshed *et al.*, 2008b).

In a work of Tan *et al.* (2008), a micro heater integrated flow focusing device was fabricated and the droplet formation was observed in the dripping regime of flow focusing device at room temperature (i.e. 25°C). As the temperature actuation starts and the temperature exceeds 30°C, the droplet formation occurs in other regimes which were previously proposed in the literature.

2.2.3.2. Pressure Actuation. The formation and the manipulation of the droplets can also be achieved by pressure actuation. Willaime *et al.* (2006) used a deflecting membrane in a microfluidics T-junction with a primary dispersed phase channel and a secondary actuation channel that connects to the main channel with a membrane. Water is filled into the actuation channel and the membrane is deflected by increasing the applied water pressure. Thus, the deflected membrane changes the instantaneous flow velocity of the dispersed phase, resulting in a change in the droplet formation frequency (forcing frequency). It is observed that the droplet formation rate increases and the droplet formation becomes regular compared to the no force applied case when the forcing frequency used as a multiple of the natural droplet formation frequency. The forcing frequency is applied within a broad range to observe its effect on the size of the droplets. It is found that there is a linear relation between the forcing frequency and the droplet size; and they are inversely

proportional to each other. Furthermore, the droplet formation becomes irregular, if the forcing frequency is not a multiple of the natural droplet formation frequency.

Chen *et al.* (2006a; 2006b) put pressure chamber on both sides of the microfluidics flow focusing orifice. The pressure chambers are activated by using pressurized gas, thus the side walls of the orifice collapse, result into droplet breakup and a change in the droplet formation frequency. Additionally, the droplet size depends on the frequency of the pressure actuation and the flow rate ratio of both dispersed and continuous phases. This method can be applied with a single actuation point (Chen *et al.*, 2006a) or simultaneously working actuation points (Chen *et al.*, 2006b).

2.2.3.3. Electro-Magnetic Field Application. The size of the primary droplets depends on both the diameter of the nozzle and the fluid properties. However, the magnitude of the applied voltage can only change the droplet size where it does not have any effect on the droplet size (Dong *et al.*, 2006a). Two types of droplets are observed in inkjet devices as primary and satellite droplets. The formation of the satellite droplets can be prevented since the voltage waveform controls their formation. Thus, only the primary droplets can be obtained regularly with a uniform size (Dong *et al.*, 2006b).

Tan *et al.* (2010) investigated the generation and manipulation of ferrofluid droplets in a micro channel with a T-junction under the influence of a magnetic field. It is found that if the magnetic field is absent, the diameter of the ferrofluid droplets decreases as the flow rate of the continuous phase increases. However, when a magnetic field is applied to the system, it is observed that the size of droplets changes with the strength of the magnetic field, magnetic field gradient and the magnetization of the ferrofluid droplets. Furthermore, the change of the shape of ferrofluid droplet is studied for the transition from the presence to the absence of the magnetic field.

2.2.3.4. Electric Field Application. Piezoelectric actuation turns the electric voltage into the mechanical energy, as the actuator vibrates the membrane is pushed and a constant amount of dispersed liquid is pinched off of the dispersed phase channel. Thus, a uniform droplet size is generated in microfluidic devices. Beulen *et al.* (2007) experimentally investigated the piezoelectric inkjet tools. Piezoelectric elements can be applied to each

channel individually (Le, 1998). The droplet generation starts only when the piezo-actuator reaches the critical voltage and exceeds this value, where the droplet formation is observed in the jetting regime (Beulen *et al.*, 2007).

Bransky *et al.* (2009) used piezoelectric actuator to generate droplets and manipulate them in microfluidics cross flow and T-junction configurations; and showed that there is no significant difference between these configurations to form droplets. Only the flow rate ratio of the dispersed phase to continuous phase has an important effect on the droplet generation. It is proved that if the flow rate ratio increases, the droplets cannot be formed individually. However, it is possible to create bursts of droplets with changing size. In the cited work, the physical mechanism of the system was studied; the droplet formation, the rate of formation and the distance between the droplets were controlled with a standard deviation of less than 0.3 %.

Gu *et al.* (2008a) put electrowetting in a micro channel with a flow focusing device to investigate the droplet formation in the presence of an electric field. Insulator covered electrodes were inserted under the micro channel. Thus, the wetting angle between the interface of immiscible liquids and the micro channel wall can be adjusted, resulting in a controlled formation of droplets. The droplet size can be arranged by controlling the electric field. The droplet diameter changes from 5 to 50 μm with changing the applied voltage. Also, the droplet formation rate increases for high electrowetting values (Gue *et al.*, 2008a; 2008b)

Three regimes are observed in flow focusing devices as dripping, tip-streaming and conical spray (Gue *et al.*, 2008a). In the conical spray regime, finite electrostatic charges accumulate on the surfaces of the droplets, which repel each other. Similarly, Kim *et al.* (2007) and He *et al.* (2010) observed the droplet formation in a flow focusing device with electrowetting property and found that the droplet size decreases as the applied voltage increases.

AC electric field was used by Tsai *et al.* (2012) to produce mono dispersed droplets in femtoliter scale. The relations of the droplet volume with the electric field and the

electric frequency were investigated. It is possible to obtain very small droplets by increasing the frequency and the AC electric field up to 1 kHz and 1 V/ μm , respectively.

Kim *et al.* (2006) worked with electrohydrodynamic mechanism to produce droplets in PDMS micro channels. They restrained the Taylor cone that is observed at electrospray by controlling the surface wetting property of the PDMS microfluidic device and also the surface tension of the liquids that used in the experiments.

Normal electric field was applied to produce droplets from immiscible fluids flowing in a micro channel (Ozen *et al.*, 2006). When the normal electric field is applied, the interface between the fluids becomes deflected and the droplet formation occurs. However, the droplets stay in a deformed shape until they exit the applied electric field region of the micro channel. Additionally, the droplet size can be controlled by adjusting the flow rate of the liquids.

2.3. Electrohydrodynamic Instabilities

Electrohydrodynamics (EHD) deals with the flow field in the presence of an electric field. The electrohydrodynamic instability of the interface between two fluids under the effect of an electric field has been studied both experimentally and numerically (Rayleigh, 1882; Melcher, 1963; Melcher and Schwarz, 1968; Kath and Hobur, 1977; Saville, 1997). Lately, the electrohydrodynamic instability has been applied to microfluidics sytems (Goranovic, 2003; Lin *et al.*, 2004; Ozen *et al.*, 2006; Uguz and Aubry, 2008). For miscible liquids, electrohydrodynamic instability can significantly enhance the mixing in micro devices at low Reynolds numbers. For immiscible liquids, the electric field may yield an interfacial instability, which in turn may generate droplets of one phase in another one (Ozen *et al.*, 2006).

Ozen *et al.* (2006) used the surface coupled model and numerically investigated the interface between two leaky dielectric fluids in the presence of normal electric field. In the surface coupled model, a jump in the electrical conductivity of the fluids exists. In this model, the electrical conductivity of each fluid is constant and there is no electrical body force. This model is discussed in detail in Section 2.3.1.

For fast charge relaxation time, i.e. large ratio of fluid time scale to electric time scale, the electrohydrodynamic equations can be simplified (Thaokar and Kumaran, 2005; Craster and Matar, 2005; Ozen *et al.*, 2006b; Uguz *et al.*, 2008) and an analytical solution is possible (Ozen *et al.*, 2006c, Uguz *et al.*, 2008). For fast charge relaxation time it is possible to understand the effect of the electric field through electrical conductivity versus permittivity ratios graph.

Linear stability analysis of the interface between two immiscible fluids in the presence of an electric field is done by Ozen *et al.* (2006) and Uguz and Aubry (2008). Thaokar and Kumaran (2005) numerically performed both linear and nonlinear analysis for dielectric fluids. Shankar and Sharma (2004) studied the linear stability analysis for either two leaky dielectrics or for two perfect dielectrics. They examined the effect of the viscosity on the growth rate. Baygents and Baldessari (1998) also studied the linear stability analysis of highly conductive electrolyte solutions under the effect of a parallel electric field. They studied the effect of the electric field in a fluid with linearly changing electrical conductivity with respect to the height. Pease and Russel (2002) compared the experimental and the analytical results of the potential difference required to destabilize a thin film. They concluded that the experimental results agree with the results of the linear stability analysis.

The difference of the electrokinetic instability from the electrohydrodynamic instability is explained in Melcher and Taylor (1969) and Saville (1997). Electrokinetic instability was first detected during the electrokinetic applications in the laboratory (Ramsey, 2001; Dang *et al.*, 2003). Furthermore, it has been studied to achieve the mixing in micro scale devices at low Reynolds number (El Moctar, 2003; Oddy *et al.*, 2001). To understand the mechanism of the electrokinetic instability, Chen and Santiago (2003) found that the conductivity gradient of the liquids is very important for the instability mechanism and the large electric field strength causes instability. This is explained by the bulk coupled model. In this model, when an electric field is applied, a separation in charge occurs in the bulk fluid. As a result of this, the charge density causes the formation of the electrical body force which is the driving force of the electrokinetic instability.

In a later work of Santiago's research group, the mechanism of the electrokinetic instability was experimentally investigated by considering some parameters such as applied electric field ratio, conductivity ratio and theoretically applied electric field (Posner and Santiago, 2006). They combined their work with the previously studied electrohydrodynamic instability works. (Hoburg and Melcher, 1976; Hoburg and Melcher, 1977; Baygents and Baldessari, 1998).

2.3.1. The Leaky Dielectric Model

The leaky dielectric model is developed by Taylor and Melcher (1969) and can predict the distortion of a Newtonian droplet under the effect of an electric field. The leaky dielectric model contains the Stokes equations to describe the motion of the fluid and defines the fluid motion by ohmic conductivity and permittivity. When an electric field is applied normal to the free surface of a perfect conductor, an instability is observed at the fluid's surface. A perfect conductor has finite conductivity where perfect dielectrics have zero conductivity. In the leaky dielectric model, the fluid is assumed to be a weak conductor and there is no bulk charge density. However, it is possible to obtain a surface charge density at the interface where the discontinuities exist in both the electrical conductivity and the permittivity. In this situation if an electric field is applied to the interface, a tangential shear stress is generated on the droplet interface (Taylor and Melcher, 1969; Saville, 1997).

The effect of electric the field on the stability of fluid-fluid interfaces has been an extensive research area since Taylor and McEwan (1965) and has been studied by many groups (Melcher and Smith, 1969; Saville, 1997; Pease and Russel, 2002; Shankar and Sharma, 2004; Craster and Matar, 2005; Thaokar and Kumaran, 2005; Ozen *et al.*, 2006; Uguz and Aubry, 2008).

These works focused on determining the dependence of the perfect dielectric or leaky dielectric systems on the critical voltage. If there is a small amount of surface charges on the interface, the interface can be destabilized. Ozen *et al.* (2006) and Sayed *et al.* (2011) investigated the interfacial instability of two leaky dielectric fluids, and Wu and Cho (2005) performed a linear stability analysis of a system that includes leaky dielectric

viscoelastic fluids. In a work of Supeene *et al.* (2005), a finite element analysis was performed for both leaky dielectric and perfect dielectric models. Li *et al.* (2007) showed that the leaky dielectric systems could be destabilized by an electric field application depending on the ratios of fluid conductivities and permittivities while perfect dielectrics can always be destabilized by the electric field. Thaokar and Kumar (2005) investigated both perfect dielectric and leaky dielectric systems; according to the results perfect dielectric systems require less critical voltage to be destabilized compared to the leaky dielectric system.

Schäffer *et al.* (2001), Lin *et al.* (2001), Pease and Russel (2002) and Herminghaus (1999) focused on the analysis of the characteristics of the linear stability of perfect dielectrics. In the same work of Pease and Russel (2002), same analysis was also done with the leaky dielectric model and they tried to understand the relation of the growth rate and the characteristic wavelength with the film thickness and the electrical conductivity. The results showed that the growth exponent and the wavelength of the system with the leaky dielectric model are much larger than the system with the perfect dielectric model.

2.3.2. Direction of the Applied Electric Field

The electric potential could be applied to achieve mixing for miscible fluids and formation of droplets for immiscible fluids. Electric field application can cause into an interfacial instability in a micro channel and it could be applied either parallel or normal to the interface of the fluids.

Patankar (2011) studied the effect of a parallel electric field on the instability of an interface between two miscible liquids with same mechanical and different electrical properties. Hsu and Patankar (2014) examined the differences between the parallel and the normal electric field applications. They showed that the normal electric field system is more unstable than the parallel electric field case and they explained this result by the larger destabilizing electric force caused by the greater asymmetry that exists in the normal electric field.

Li *et al.* (2011) experimentally and analytically investigated the instability of the interface between two immiscible fluids with different electrical properties when it is subjected to a normal electric field. They found that the normal electric can have a destabilizing effect, and the increase in the flow rate and the viscosity have destabilizing effect. They also showed that the experimental results successfully match the analytical findings. Similarly Uguz *et al.* (2008) worked with immiscible fluids and investigated the effects of both parallel and normal electric fields. They provided a stability diagram of electrical conductivity and permittivity ratios showing regions where parallel and normal electric fields have destabilizing effect. In a further work (Uguz and Aubry, 2008), they showed the growth rate-wavelength relation and the neutral stability curves. Abdella and Rasmussen (1997), Ozen *et al.* (2006) also studied the immiscible fluids and the normal electric field effect. They experimentally showed that monodisperse droplets can be formed by applying a normal electric field. Ersoy (2011) and Nurocak (2012) investigated the effect of the direction of the electric field on the interfacial instability between a Newtonian and a viscoelastic phase.

2.3.3. Direct Current or Alternating Current

The electricity can flow in two ways i.e. direct or alternating. In DC field the electrons flow in a single direction, where in AC field they change their directions and flow forward and backward.

Working with DC field is more common in the literature (Shankar and Sharma, 2004; Goranovic, 2005; Ozen *et al.*, 2006a; El-Sayed *et al.*, 2011) than the AC field to investigate electrohydrodynamic instabilities. Some of the works with the AC field are done by Robert and Kumar (2009), Rout and Choudhory (2006), Gambhine and Thaokar (2010; 2011) and Espin *et al.* (2013).

Ozen *et al.* (2006a) applied DC electric field perpendicularly to the interface between immiscible liquid to generate monodisperse droplets and controlled their size. Kim *et al.* (2007) investigated the droplet formation in a micro device with flow focusing geometry under the effect on a DC electric field. They studied the dependence of the droplet size on the applied potential difference for changing flow rate ratios. Uguz and Aubry (2008)

analyzed the electrohydrodynamic interfacial instability of two leaky dielectrics in the presence of normal and parallel DC electric fields. Choi *et al.* (2011) controlled the volume of the generated droplet by the applied DC electric field.

3. EXPERIMENTAL WORK

3.1. Chemicals

A list of the chemicals used in this study is given in Table 3.1.

Table 3.1 The chemicals used in the experiments.

Chemicals	Formula	Source
Ethylene Glycol	$C_2H_6O_2$	Merck
Silicone Oil 5	$(C_2H_6OSi)_n$	Brookfield
Silicone Oil 10	$(C_2H_6OSi)_n$	Brookfield
Silicone Oil 20	$(C_2H_6OSi)_n$	Sigma-Aldrich
Silicone Oil 50	$(C_2H_6OSi)_n$	Brookfield
Silicone Oil 100	$(C_2H_6OSi)_n$	Brookfield
Castor Oil	$C_{57}H_{104}O_9$	Sankim
Olive Oil	-	Komili

Table 3.2. lists the physical properties of the chemicals used in the experiments. These properties are density, kinematic viscosity, conductivity, and relative permittivity of the liquids at 25°C. The interfacial tension, γ between ethylene glycol and silicone oil is reported to be 18 mN/m. (Bico *et al.*, 2000). The physical properties of ethylene glycol are obtained from MEGlobal (2008). The density, viscosity and relative permittivity data of silicone oils are found in the technical data sheet of Shin Etsu Co. (2004). The conductivities of silicone oils are given in Zhag *et al.* (2006). Relative permittivities and conductivities of castor and silicone oils are reported in a work of Paranjep *et al.* (1935) and Saied *et al.* (2004), respectively. Finally the density and viscosity of castor oil and olive oil can be found in the technical data sheet of Shri Ram Co. (2014) and in a study of Fasina *et al.* (2008), respectively.

Table 3.1. Physical properties of the chemicals used in the experiments.

Chemicals	Density, ρ (kg/m ³)	Kinematic Viscosity, ν (cSt)	Conductivity, σ (S/m)	Relative Permittivity, ϵ
Ethylene Glycol	1110	15	1.1E-04	37.70
Silicone Oil 5	915	5	1.0E-13	2.60
Silicone Oil 10	930	10	1.0E-13	2.65
Silicone Oil 20	950	20	1.0E-13	2.70
Silicone Oil 50	960	50	1.0E-13	2.72
Silicone Oil 100	965	100	1.0E-13	2.74
Castor Oil	961	1025	5.0E-12	4.78
Olive Oil	911	92	3.0E-11	3.35

3.2. Experimental Set-up

In this section, the experimental set-up is explained and the geometries of the micro channels are demonstrated in detail. As shown in Figure 3.1, the experimental set-up includes a micro channel, a syringe pump, a microscope, a high speed camera and a DC source.

The liquids are pumped with a twin syringe pump (Harvard Apparatus Pump 33, Holliston, MA) and two syringes (Set Medical Inc, Istanbul, Turkey) with a diameter of 19.13 mm that are used as reservoirs for the liquids. Butterfly catheter infusion sets (Harvard Apparatus Standard Butterfly Catheter Infusion Sets, Holliston, MA) with a needle diameter of 21G (0.81 mm) are attached to the syringes and they are connected to the inlets of the micro channels by using poly ethylene micro medical tubings (Scientific Commodities Inc., Lake Havasu City, Arizona) with 0.72 mm of inner diameter (ID) and 1.22 mm of outer diameter (OD). Thus, the connection is successfully achieved from syringes to the inlets of the micro channel and leakage is prevented.

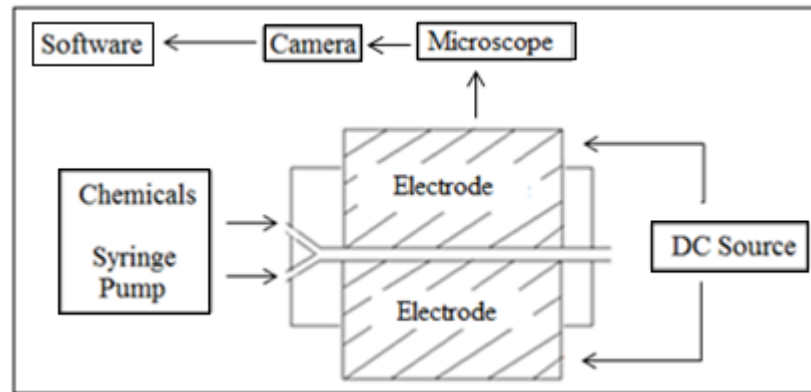


Figure 3.1. Sketch of the experimental setup.

For electric field applications, Glassman EQ Series 1200 Watt Regulated High Voltage DC supplier is connected to the electrodes of the micro channels. To check the applied voltage, a high accuracy multimeter (Extech Instruments Multimaster 570A Tru RMS) is used in case of need.

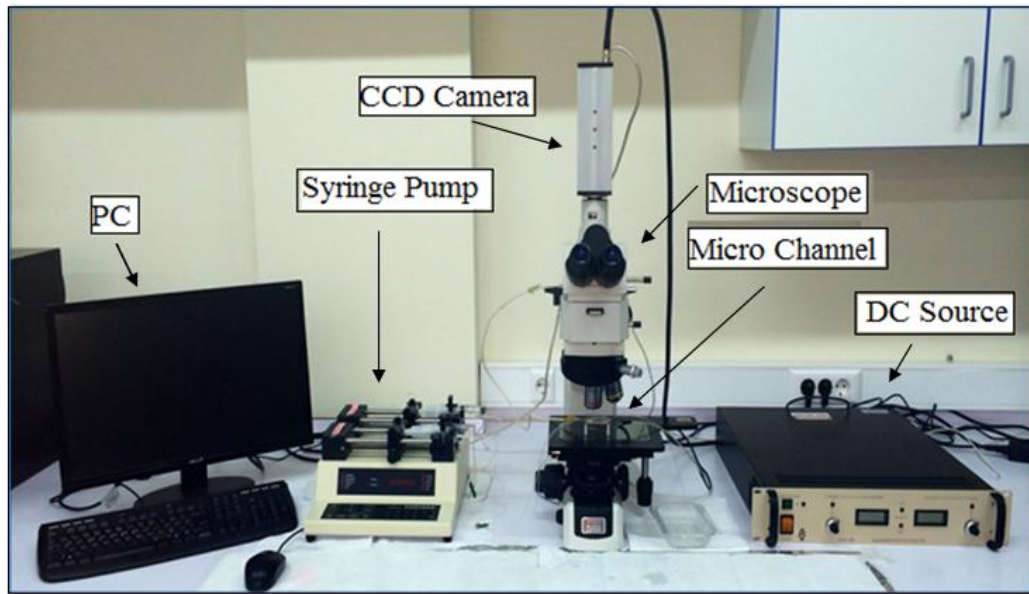


Figure 3.2. Experimental set-up.

Nikon Eclipse LV100POL Microscope with a 5x objective is used to visualize the experiments. The image is recorded up to 250 fps with Fastec InLine High-Speed Camera that is connected to a computer with the software (FIMS 3.0.5). Then, the recorded videos are played with MIDAS Player at slow speed.

Three different micro channels are fabricated by TMR Engineering, Micanopy, FL, USA, one for parallel electric field and the other two for normal electric field applications. The micro channels are of Y shapes with two stainless steel inlets of 1.5 mm outside diameter and the micro channels are made of PMMA (Plexi Glass). All of the micro channels are 50 mm long and 200 μm in deep.

For the parallel electric field applications, the micro channel is equipped with two conductive copper wires, one is located at the entrance and the other one is at the exit of the micro channel as seen in Figure 3.3c, Figure 3.4a and Figure 3.4b. The width of the micro channel for the parallel electric application is 1.0 mm and the distance between the two electrodes is 30 mm as shown in Figure 3.3c. The depth of the micro channel is 200 μm in deep.

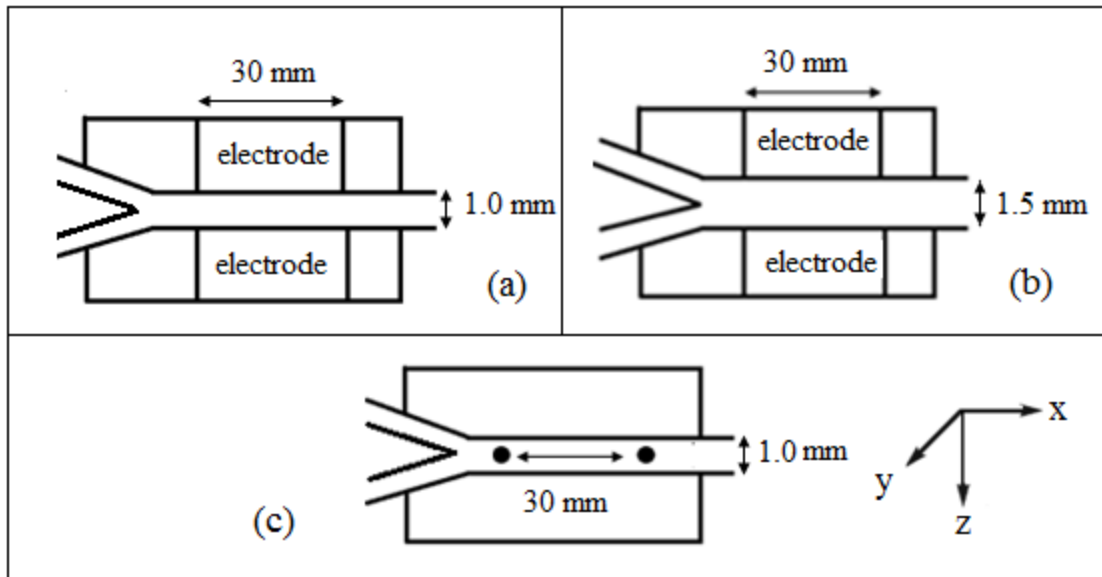


Figure 3.3. Channel geometry (a) normal electric field with a width of 1.0 mm (b) normal electric field with a width of 1.5 mm (c) parallel electric field with a width of 1.0 mm.

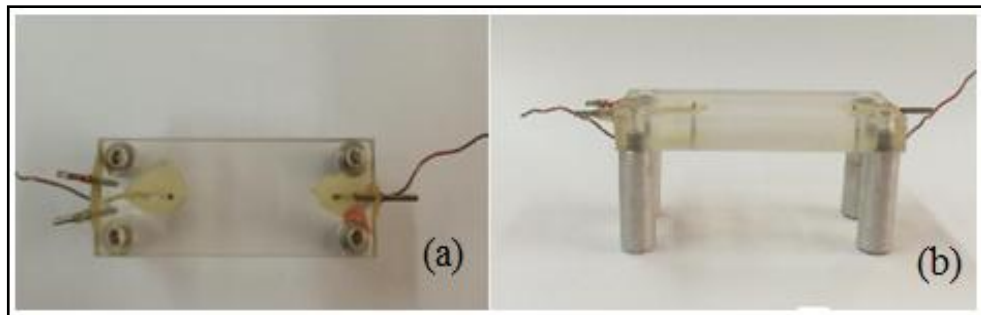


Figure 3.4. Micro channel with parallel electric field application (a) top view and (b) side view.

For the normal electric field application, two copper sheets each having length of 30 mm, width of 15 mm and thickness of 200 μm are placed facing one another as seen in Figure 3.3a and Figure 3.3b. The copper sheets are put between the top and bottom PMMA plates, so that the electrodes become the side walls of the micro channel. As a result of this design, the depth of the micro channel is actually the thickness of the copper sheets.

Two micro channels of 1.0 mm (Figure 3.3a) and 1.5 mm (Figure 3.3b) width are used for the normal electric field application to understand the effect of the width of the micro channel (the distance between the electrodes) on the stability of the interface. Figure 3.5 presents the micro channel with 1.0 mm from the top (Figure 3.5a) and from side views (Figure 3.5b).

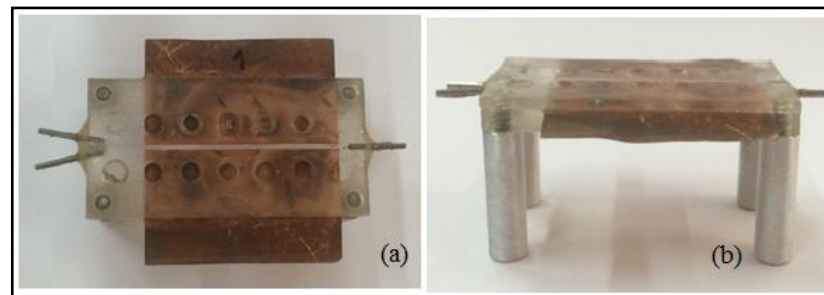


Figure 3.5. Micro channel with a width of 1.0 mm and normal electric field application
(a) top view and (b) side view.

3.3. Procedure of the Experiment

- Once the experimental set-up is ready, the liquids, which are assumed Newtonian and incompressible, are subject to a pressure gradient by the twin syringe pump and each liquid flows into the micro channel through its own inlet until a flat interface is formed between them as shown in Figure 3.7.

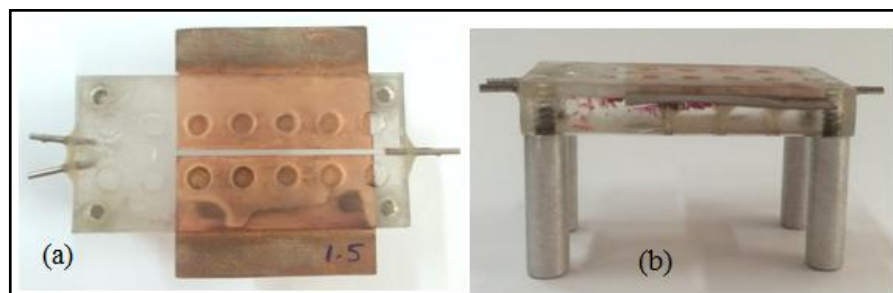


Figure 3.6. Micro channel with a width of 1.5 mm and normal electric field application
(a) top view and (b) side view.

- Once a flat interface is obtained a potential difference is applied with DC source parallel or normal to the interface depending on the micro channel used. The voltage is raised by increments of 10 V and at each step the flatness of the interface is checked.
- At some voltage, called critical voltage the interface between the two immiscible liquids start deflecting. As an example, the interface between 50 cSt Silicone Oil and Ethylene Glycol both flowing at 40 $\mu\text{L}/\text{min}$ becomes unstable, i.e. starts deflecting when the voltage reached 725 V applied normal to the interface in the channel with 1.0 mm width. The deflected interface can be seen in Figure 3.8.
- To verify the critical potential difference of the system, each experiment is repeated at least three times and the average of the results is reported.
- At the critical voltage, if the interface is observed long enough, it is possible that the amplitude of the deflection amplifies even the interface may flap one of the walls causing its rupture (Figure 3.10 and Figure 3.11).

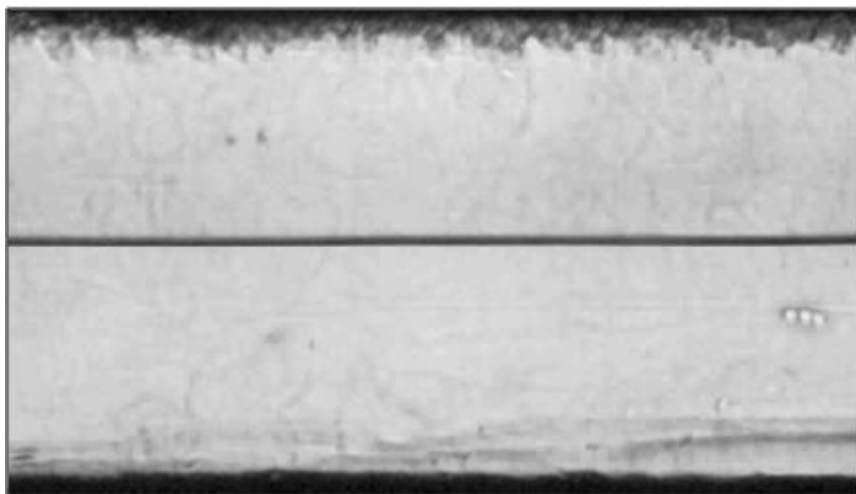


Figure 3.7. The flat interface between silicone oil, 50 cSt and ethylene glycol; both with a flow rate of 40 $\mu\text{L}/\text{min}$.

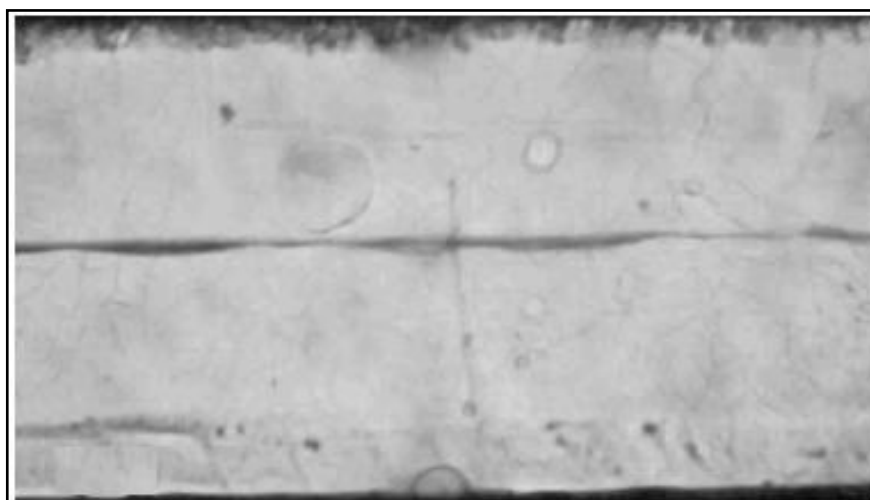


Figure 3.8. Critical voltage detected for the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of $40\mu\text{L}/\text{min}$ at 725 V.

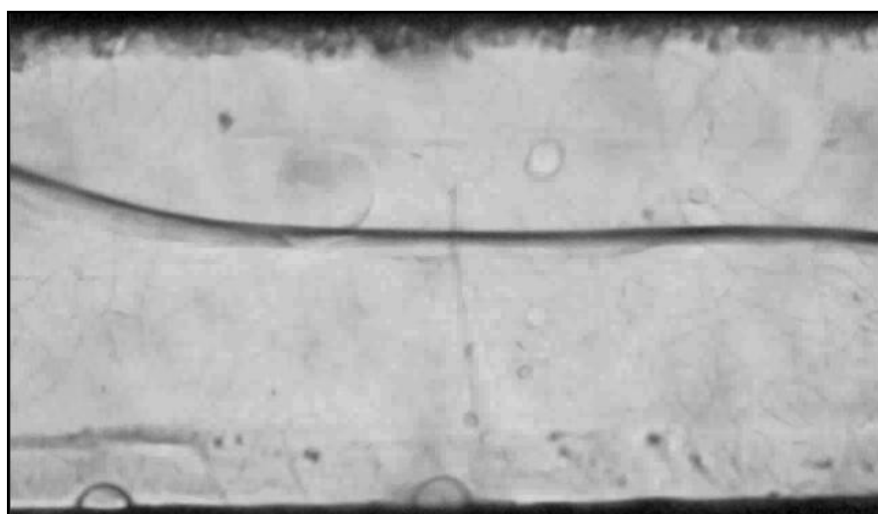


Figure 3.9. The increased instability at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of $40\mu\text{L}/\text{min}$ at 725 V.

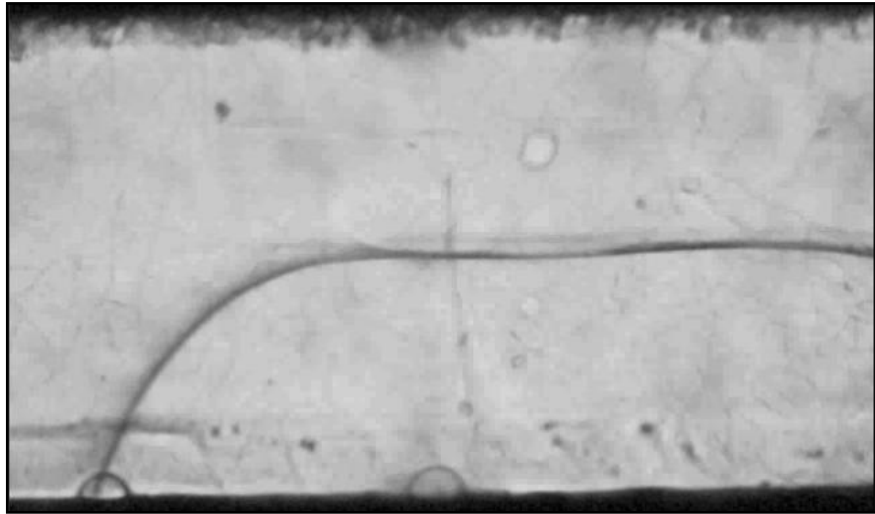


Figure 3.10. The rupture at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of $40\mu\text{L}/\text{min}$.

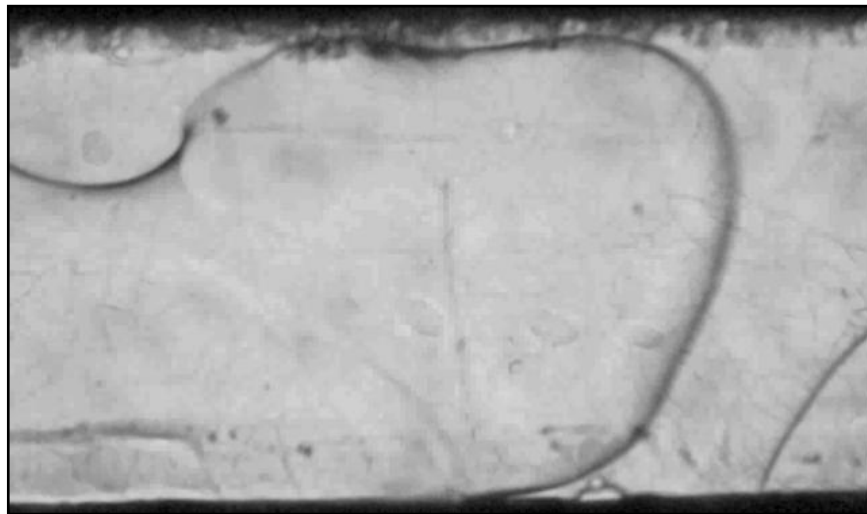


Figure 3.11. The complete rupture at the interface of silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of $40\mu\text{L}/\text{min}$.

- When the intensity of the applied potential difference is decreased slowly, the interface starts to become less unstable as shown in Figure 3.12a and Figure 12b and then it regains its flat shape as seen in Figure 3.12c.

- Finally, the interface between the immiscible liquids becomes flat again at the end of the experiment.
- When the interface is flat as shown in Figure 3.12c and the DC source is turned off, the same procedure can be done for the same liquids but with different flow rates without making any change. In addition, all of the experiments are performed by keeping the temperature at 25°C.
- To perform the experiments with different liquid combinations, all of the syringes, connection sets, and tubing are replaced. Also, the micro channels are cleaned carefully.

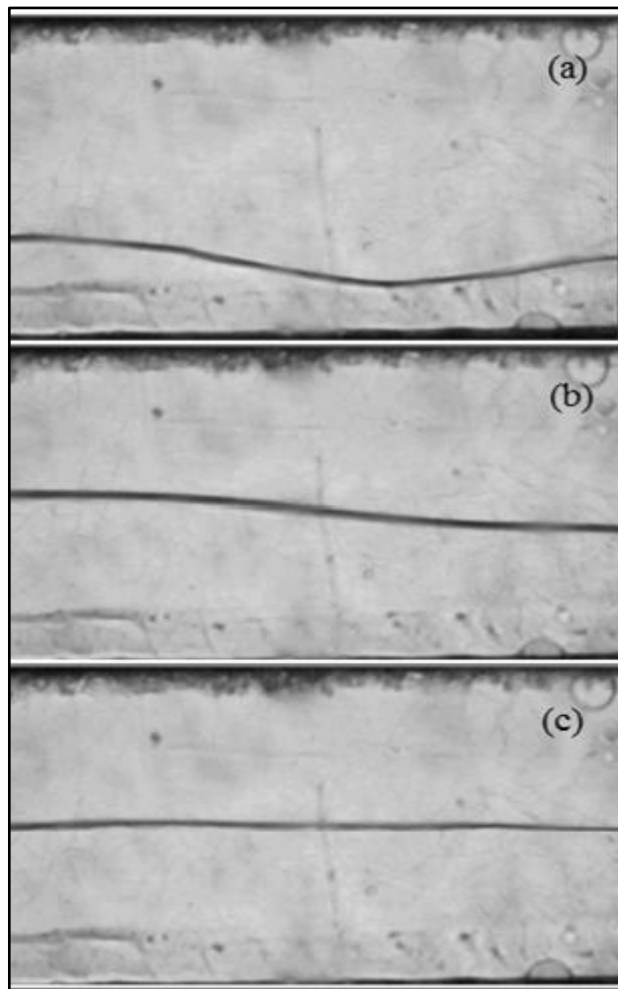


Figure 3.12. The interface of the silicone oil, 50 cSt and ethylene glycol system; both with a flow rate of 40 μ L/min the voltage is decreased below its critical value.

4. RESULTS AND DISCUSSION

In this chapter, the results of the experiments of the stability of the interface between two immiscible and Newtonian liquids in the presence of an electric field applied either parallel or normal to the flat interface are shown. First, the evolution of the interfacial instability is introduced. In this work, the effects of the four different parameters, namely viscosity and flow rate ratios of the liquids, the channel width and the direction of the electric field on the critical voltage at which the interface starts deflecting are studied.

4.1. Evolution of the Electrohydrodynamic Instability

The interface between two immiscible and Newtonian liquids can be destabilized when the liquids are subjected to an electric field. First, a flat interface is formed between the liquids in a micro channel and when an electric field is applied to the flat interface, the interface starts to deflect at the critical voltage of the system. The electric field is applied either parallel or normal to the flat interface between the liquids.

Figure 4.1 shows the deflection that occurs at the interface between ethylene glycol and 50 cSt silicone oil in the presence of a normal electric field. In Figure 4.1, the top and the bottom liquids are ethylene glycol and silicone oil, 50 cSt, respectively. The liquids flow side by side in a micro channel with a flow rate of 10 $\mu\text{L}/\text{min}$. At the beginning of the experiment, the interface is flat at time $t=0$ as seen in Figure 4.1a. Then, an electric field normal to the flat interface is increased with increments of 10 V and the interface is observed for a deflection. The system of ethylene glycol and silicone oil, 50 cSt reaches its critical potential difference of 880 V at $t = 75.1$ s (see Figure 4.1b). Hence, the interface starts to deflect at the critical potential difference (Figure 4.1c). When the system reaches its critical voltage, the magnitude of the applied potential difference is stopped increasing. The evolution of the deflection of the interface is shown from Figure 4.1b through Figure 4.1o.

If the magnitude of the deflection occurred at the interface is amplified as happened in ethylene glycol-silicone oil, 50 cSt case (see from Figure 4.1b to Figure 4.1h), the interface may flap one of the walls as seen in Figure 4.1h. If the interface flaps a wall, the interface may be ruptured as occurred in Figure 4.1j. If the applied potential difference is decreased, as at time $t=495.6\text{ s}$ (Figure 4.1o), it is possible for the interface to regain its flat shape as shown in Figure 4.1t and change to its original position (Figure 4.1t).

In this thesis, the aim is to determine the critical voltage at which the interface starts deflecting as a function of viscosity and height ratios of liquids, the channel width and the direction of the electric field.

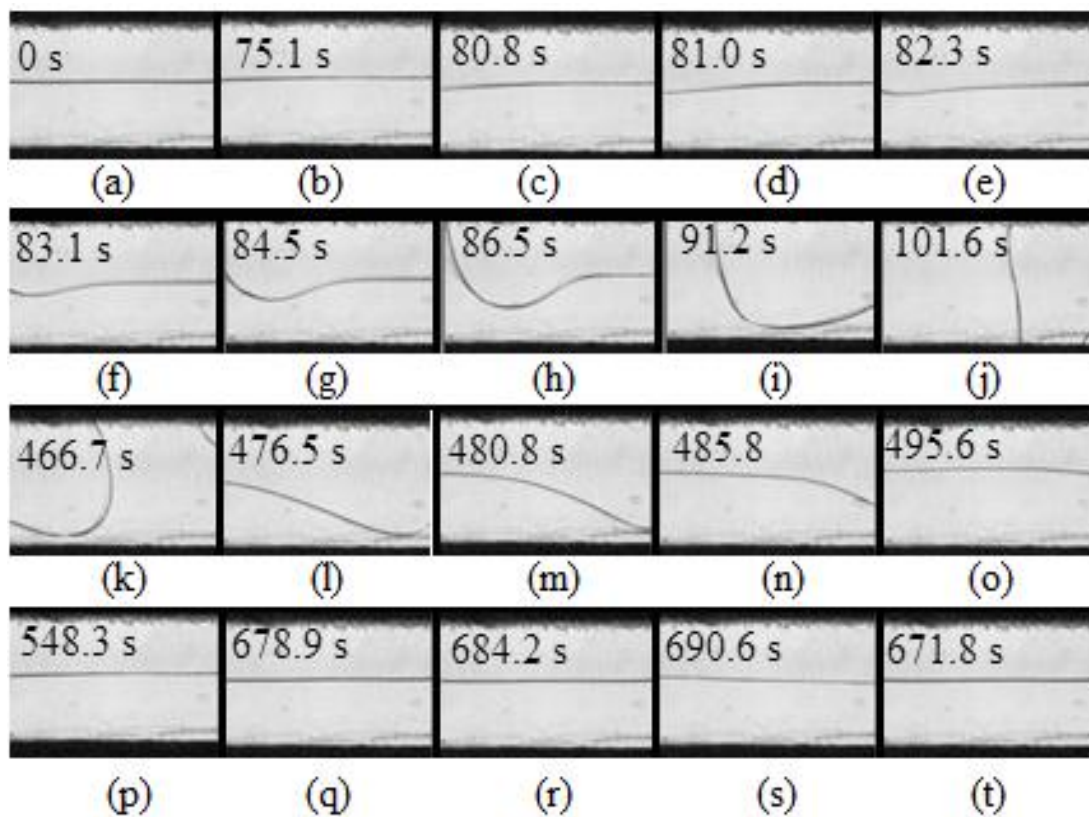


Figure 4.1. The instability occurred at the interface between ethylene glycol and silicone oil, 50 cSt at a critical potential difference of 880 V with respect to time in the presence of a normal electric field.

4.2. The Effect of the Viscosity Ratio

In this section, the effect of the viscosity ratio on the stability of the interface between immiscible liquids under the influence of normal electric field is analyzed. The experiments are carried in micro channels with different widths.

In the experiments, ethylene glycol is paired with silicone oils of different viscosities to investigate the effect of the viscosity ratio. Thus, the viscosity ratio of ethylene glycol and silicone oil systems is changed from 0.033 to 6.667. The viscosity ratio, μ_r is the ratio of the viscosity of the first liquid $\mu^{(1)}$ silicone oil to the viscosity of the second liquid, $\mu^{(2)}$ ethylene glycol.

Figures 4.2 and Figure 4.2b show the effect of the viscosity ratio on the critical potential difference (V) needed to destabilize ethylene glycol - silicone oil systems in the presence of a normal electric field in a micro channel with a width of 1 mm and 1.5 mm, respectively. The results shown in Figures 4.2a and b are for a fixed flow rate ratio, Q_r of 1.5.

The experimental results show that for a fixed flow rate ratio and a channel width, the critical potential difference needed to destabilize the flat interface increases with increasing viscosity ratio under the effect of a normal electric field. The results indicate that a system requires higher potential difference to be destabilized as the viscosity ratio of that system increases. This result means that the increase in the viscosity of the silicone oil has a stabilizing effect on the on the interface between the immiscible liquids in the presence of a normal electric field.

The results of the experiments can be shown in dimensionless form using the electric number, E_b . The electric number is a dimensionless parameter and it is defined by Uguz *et al.* (2008) as;

$$E_b = \frac{\epsilon_0 V^2}{\mu^{(2)} H^{(2)} U} \quad (4.1)$$

where ϵ_0 , $H^{(2)}$, U and V are the vacuum permittivity, height of the liquid of the 2nd phase, the characteristic velocity that is defined as $\gamma/\mu^{(2)}$ and the applied base voltage, respectively. Here γ is the interfacial tension between ethylene glycol and silicone oil.

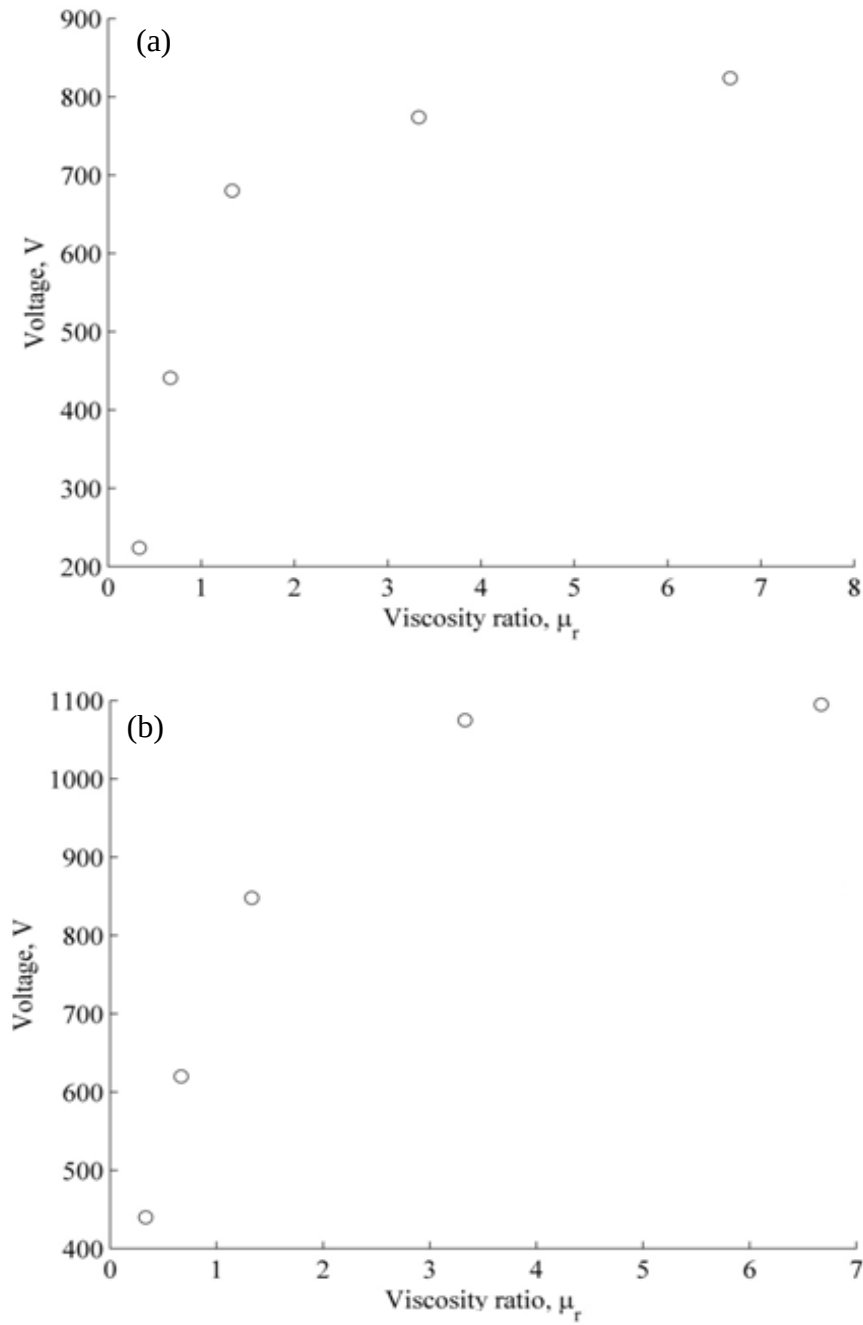


Figure 4.2. The effect of the viscosity ratio on the voltage in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b).

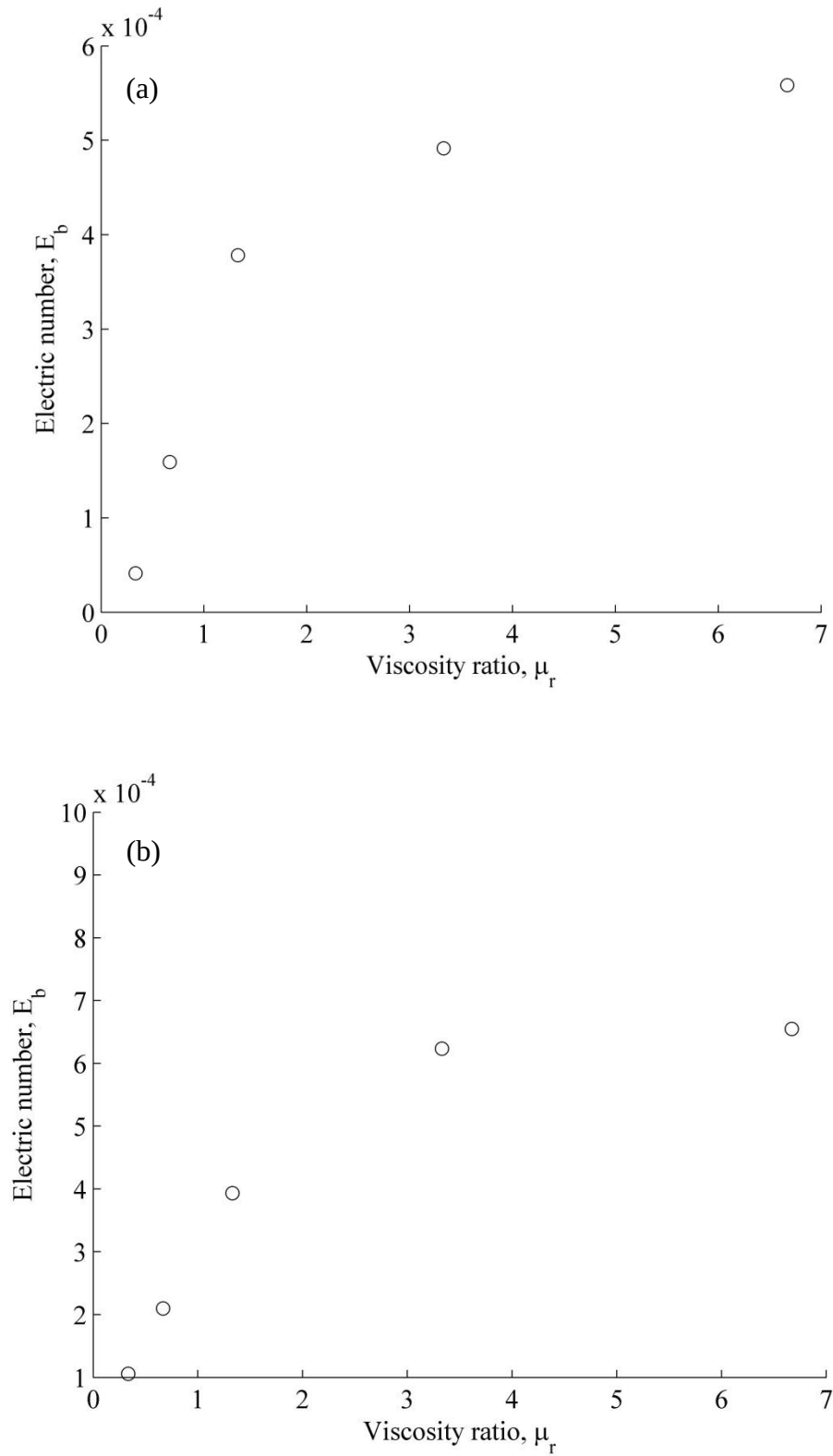


Figure 4.3. The effect of the viscosity ratio on the electric number, E_b in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b).

In Figures 4.3a and 4.3b the effect of the viscosity ratio on the dimensionless electric number, E_b is presented for a micro channel with a width of 1.0 mm and 1.5 mm, respectively. As seen from Figure 4.5a and Figure 4.5b, while the viscosity ratio of a system increases, the dimensionless electric number, E_b of the system also increases.

4.3. The Effect of the Flow Rate Ratio

4.3.1. The Effect of the Changing Flow Rate Ratio

In this section, the results of the experiments to investigate the effect of the flow rate ratio on the stability of the interface between immiscible liquids in the presence of a normal applied electric field are given. The flow rate ratio, Q_r is the ratio of the flow of the first liquid, $Q^{(1)}$ to the flow rate of the second liquid, $Q^{(2)}$.

In the experiments, the investigated flow rate ratios, Q_r are arranged by changing the flow rates of silicone oil, $Q^{(1)}$ and ethylene glycol, $Q^{(2)}$ in such a way the flow rate ratios of the systems become 0.50, 0.67, 1.00, 1.50 and 2.00. The experiments are performed for silicone oils of different viscosities, i.e. 5 cSt, 10 cSt, 20 cSt, 50 cSt and 100 cSt.

In Figure 4.4, the potential difference (V) required to destabilize ethylene glycol – silicone oil interface with different flow rate ratios in the presence of a normal electric field is shown. The micro channels used in the experiments have widths of 1.0 mm (Figure 4.6a) and 1.5 mm (Figure 4.6b). The systems with different viscosity ratios were presented in Section 4.2 and they change from 0.033 to 6.667. As seen from Figure 4.4, increasing the flow rate ratio of the system with a fixed viscosity ratio and a channel width has a stabilizing effect on the ethylene glycol – silicone oil interface. In other words, as the flow rate of the silicone oil, $Q^{(1)}$ is increased for a fixed flow rate of ethylene glycol, $Q^{(2)}$, the flow rate ratio, Q_r of the system increases. Thus, the interface between ethylene glycol and silicone oil becomes more stable compared to the case having a lower flow rate ratio, Q_r .

In Figure 4.5, the effect of the flow rate ratio on the stability of the silicone oil and ethylene glycol systems is shown in terms of the dimensionless electric number, E_b in the presence of a normal electric field for micro channel widths of 1.0 mm (Figure 4.5a) and

1.5 mm (Figure 4.5b). The results show that the electric number, E_b increases as the flow rate ratio of the ethylene glycol – silicone oil system increases.

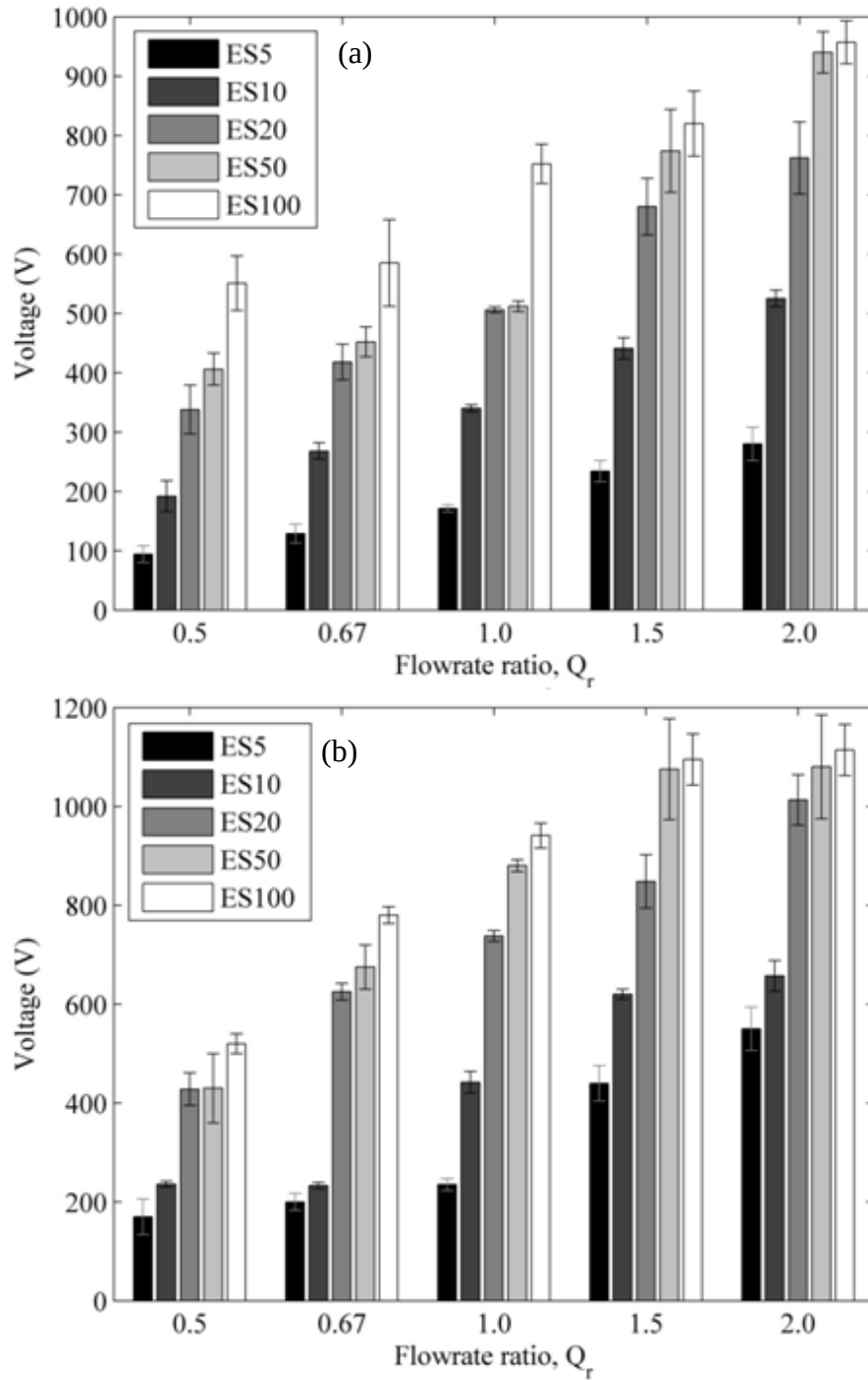


Figure 4.4. Effect of the flow rate ratio on the potential difference in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b).

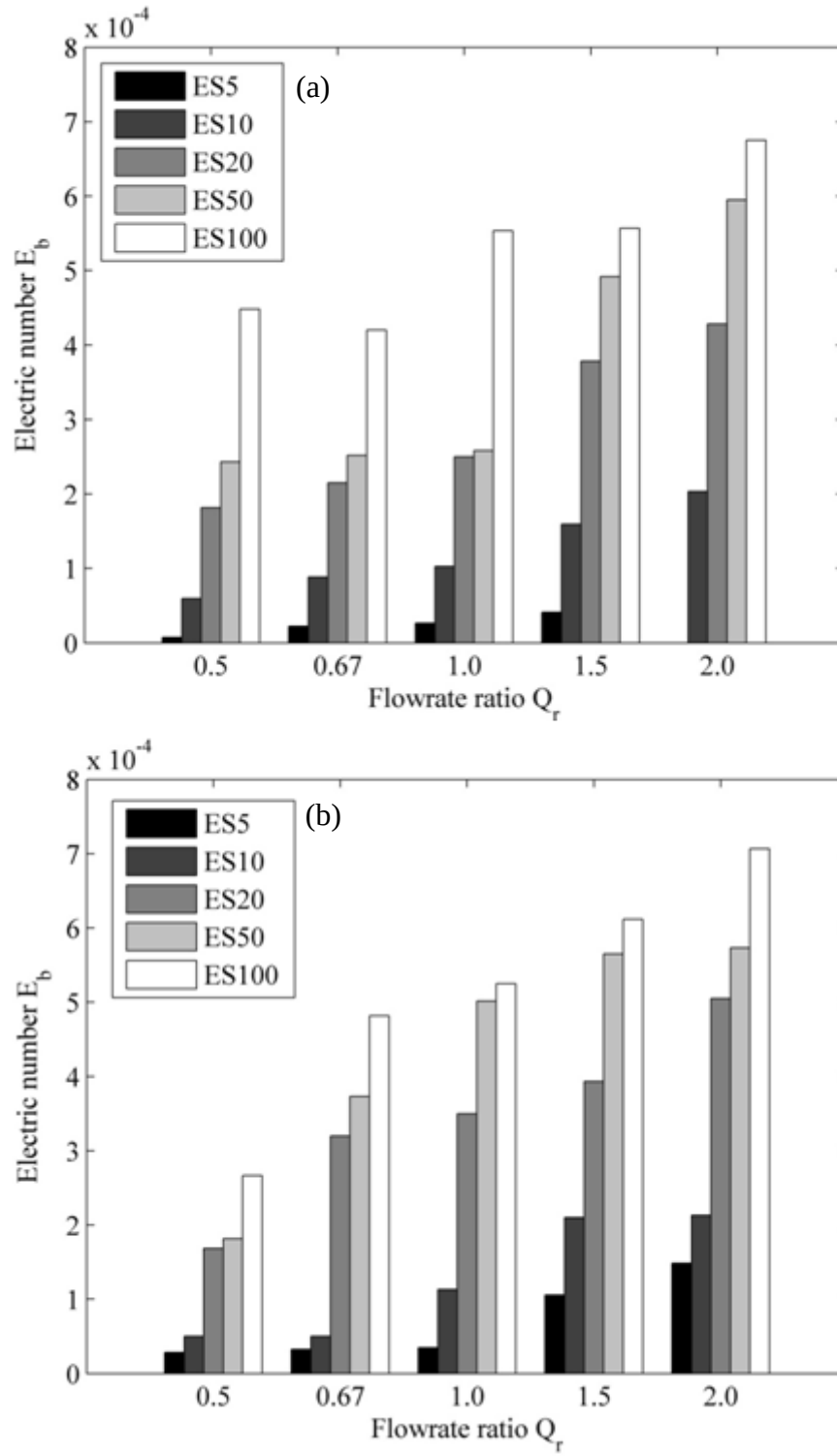


Figure 4.5. The effect of the flow rate ratio on the electric number in the presence of a normal electric field for the micro channels with 1.0 mm (a) and 1.5 mm widths (b).

4.3.2. The Effect of Constant Flow Rate Ratio

In this section, a special case of the effect of the flow rate ratio is investigated. In this case, the flow rates of a system are changed but the ratios of the flow rates are kept constant at unity. In the experiments, the flow rates are altered from 10/10 $\mu\text{L}/\text{min}$ to 50/50 $\mu\text{L}/\text{min}$ with increments of 10 $\mu\text{L}/\text{min}$ and even the stationary liquids case is included as 0/0.

Figure 4.6 shows the potential difference needed to destabilize the flat interface between ethylene glycol and silicone oil pairs. The experiments are performed for each ethylene glycol-silicone oil system with different viscosity ratios. Normal electric field is applied to the flat interface while the liquids flow with the same flow rates. As seen from Figure 4.6, if the flow rate ratio of a system is constant at unity even when the flow rates are changed, the interface between two immiscible liquids requires same potential difference (V) to destabilize. This result shows that the flow rate ratio, Q_r is the important parameter for a system to determine the critical potential difference, not the individual flow rates of the liquids.

In addition to the experiments with constant flow rate ratios, no-flow case is also investigated. No-flow case is achieved when the system reaches the quiescent state after stopping the flows of the liquids. When the flow is stopped, the height of each layer becomes equal each other, so that the height ratio, H_r of the system becomes unity. Normal electric field is applied to the flat interface and it is observed that the interface starts deflecting at the same potential difference (see 0/0 case in Figure 4.6) with the flow case. If there is a flow in the micro channel, the destabilized interface can regain its flat shape when the applied electric field is decreased and turned off. However, the destabilized interface cannot regain its flat shape for the no-flow case.

In Figure 4.6b, the effect of constant flow rate ratio is also shown in terms of the dimensionless electric number, E_b for ethylene glycol – silicone oil pairs. Like the potential difference, the dimensionless electric number is same for constant flow ratio at no - flow cases.

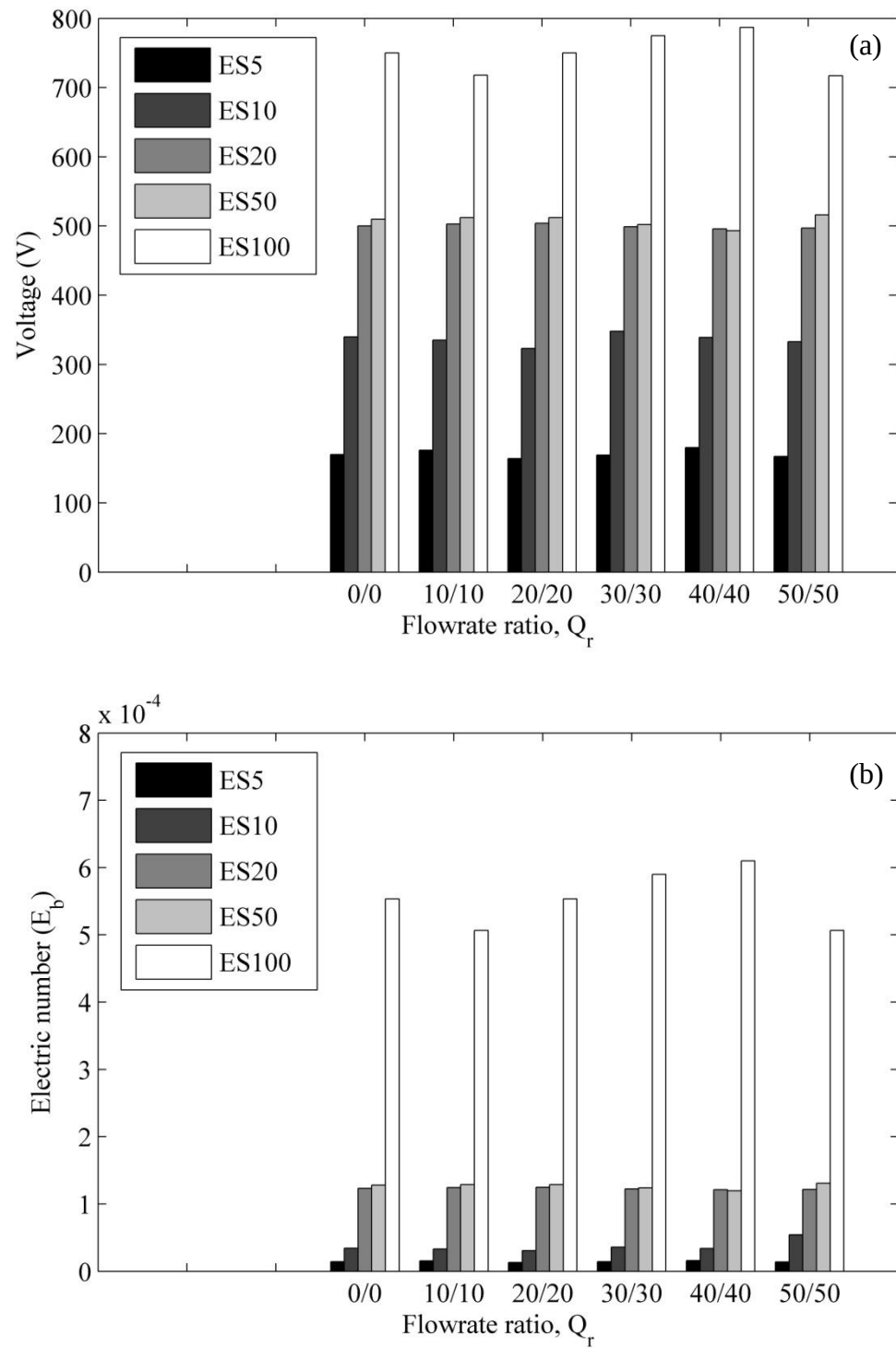


Figure 4.6. The effect of the same flow rate ratio of unity for silicone oil-ethylene glycol pairs on the voltage (a) and electric number, E_b (b) in the presence of a normal electric field.

4.4. The Effect of the Channel Width

Two micro channels of 1.0 mm and 1.5 mm widths are used for the normal electric field applications to understand the effect of the width of the micro channel on the stability of the interface. Thus, the distance between the electrodes becomes 1.0 mm and 1.5 mm. In this section, the experimental results of the ethylene glycol – silicone oil pairs are compared for different channel widths.

The experiments are performed for two different channel widths in the presence of a normal electric field and they are repeated for all ethylene glycol – silicone oil pairs. The critical potential difference needed to destabilize each system is recorded and the results are shown in Figure 4.7 for a fixed flow rate ratio of unity where the individual flow rates are 10 $\mu\text{L}/\text{min}$. It can be seen from Figure 4.8 that as the width of the channel increases, the critical potential difference needed to destabilize the interface also increases.

Figure 4.7b presents the electric number, E_b of each system when the system is destabilized at its critical potential difference. Increasing the width of the micro channel results in an increase in the electric number, E_b of the system.

4.5. The Effect of the Direction of the Electric Field

As the last parameter, the effect of the direction of the electric field is investigated. The electric field is applied either normal or parallel to the flat interface formed between the two immiscible liquids where the width of the micro channel is fixed at 1.0 mm.

The liquid pairs of ethylene glycol – silicone oils, castor oil – silicone oils and olive oil – silicone oils are investigated under the effect of parallel applied electric field. The combinations ethylene glycol – silicone oils are also studied in the presence of normal electric field and their results are already presented in the previous sections.

In the parallel applied electric field experiments, each liquid pair is studied for different flow rate ratios, Q_r that are 0.5, 0.67, 1.0, 1.5 and 2.0. A maximum of 6,000 V corresponding to 6×10^6 V/m electric field is applied to each system and it is observed that

the parallel electric field does not have a destabilizing effect on any of the systems. This was an expected result for the parallel applied electric field experiments since according to the stability diagram provided by Uguz *et al.* (2008), all of the liquid pairs that are studied in this work are in a region where only a normal electric field has a destabilizing effect on the interface.

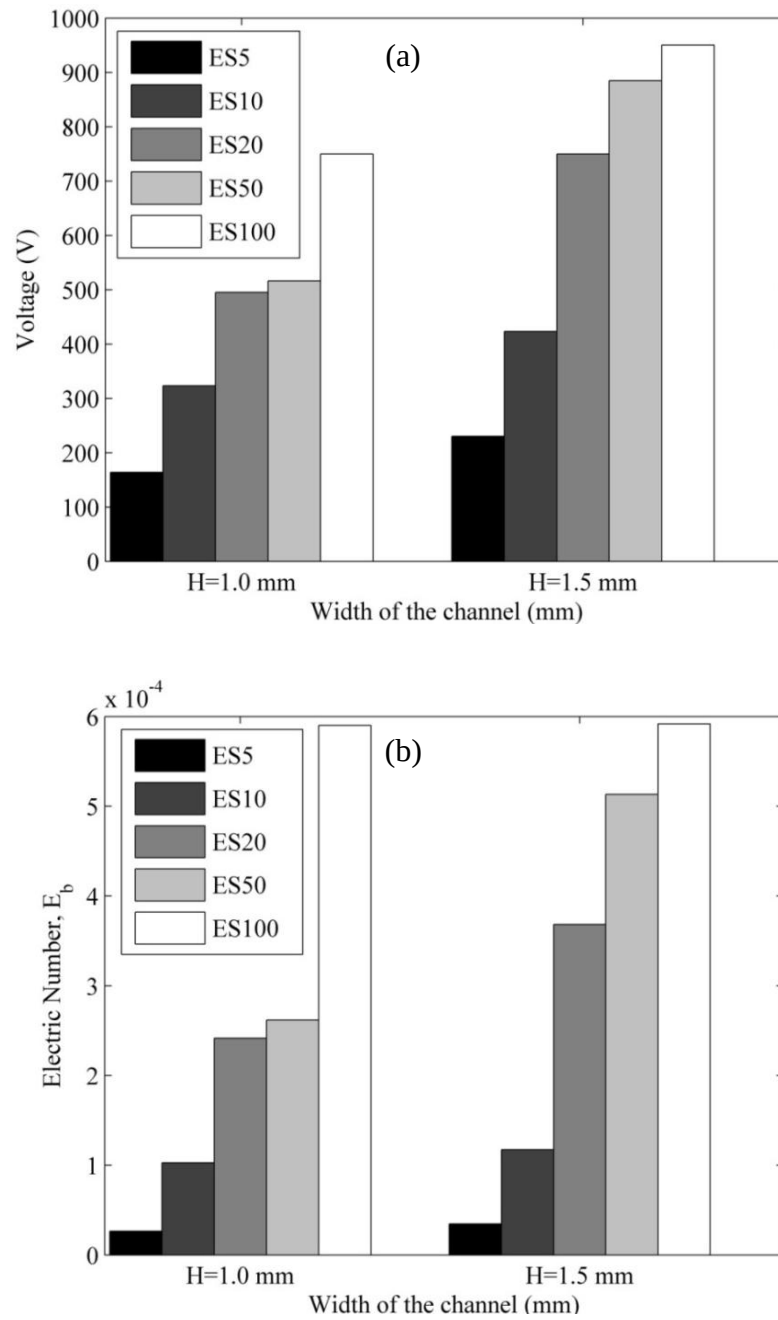


Figure 4.7. The effect of the width of the channel for silicone oil-ethylene glycol pairs on the voltage (a) and electric number, E_b (b) in the presence of a normal electric field.

5. CONCLUSIONS AND RECOMMENDATIONS

5.1. Conclusions

The electrohydrodynamic instability of the interface between two immiscible liquids with different electrical properties flowing in a PMMA micro channel is studied experimentally in the presence of an electric field. The electric field is applied either parallel or normal to the flat interface between these liquids. Each experiment is repeated at least three times and it is observed that results are in good agreement.

The critical potential difference needed to destabilize the flat interface between immiscible liquids is investigated for several parameters. The ratios of the viscosities and flow rates of the liquids, the width of the micro channel and the direction of the applied electric field are studied in the experiments. The results are also presented in a dimensionless form using the ratios of the parameters and the electric number, E_b .

Ethylene glycol and silicone oils of different viscosities are used to investigate the effect of the viscosity ratio of the liquids on the stability of the interface in the presence of a normal electric field. It is observed that the increase in the viscosity of the silicone oil has a stabilizing effect on the on the interface between immiscible liquids in the presence of a normal electric field.

Next, the effect of the flow rate on the stability is analyzed. It is found that the potential difference needed to destabilize the flat interface between the immiscible liquids increases with increasing flow rate ratio. Hence, the interface becomes more stable as the ratio of the flow rates increases. A special case of the flow rate ratio is studied where the flow rate ratio is kept constant at unity. According to the results, it is concluded that the important parameter for a system is the flow rate ratio, not the individual flow rates. Even if the flow rates are changed, a system destabilizes at the same critical potential difference as long as the ratio of the flow rates is kept constant. In addition, the critical potential difference remains same for the stationary flow case.

Moreover, the effect of the channel width is studied under the influence of a normal electric field. As a result, it is found that as the width of the channel increases, the critical potential difference needed to destabilize the interface also increases.

Finally, a parallel electric field is applied to the flat interface formed between the immiscible liquids to compare the results with the normal electric field case. The ranges of voltages needed to destabilize the interface in the normal electric field applied experiments are in between 94 and 1090 volts for ethylene glycol-silicone oil systems. The ranges of voltages needed to destabilize the interface in the normal electric field applied experiments are in between 94 and 1090 volts for the ethylene glycol-silicone oil systems. A maximum of 6000 volts is applied in the parallel electric field experiments and it is observed that the parallel electric field has no destabilizing effect on the ethylene glycol-silicone oils, castor oil-silicone oils and olive oil-silicone oils systems. The results of the parallel electric field experiments are in an agreement with the literature (Uguz and Aubry, 2008) since the liquid combinations used in the experiments are in a region where only normal electric field has a stabilizing effect on the interface due to the conductivity and permittivity ratios of the liquids.

5.2. Recommendations

In this thesis, our aim was to investigate the electrohydrodynamic instability of the interface between different liquid pairs for different parameters; including the ratios of the flow rates and viscosities, the effect of the channel width and the direction of the applied electric field.

The instability of the interface between castor oil/silicone oils and olive oil/silicone oils liquid pairs can be studied experimentally in the presence of a normal electric field. The results of these experiments can be compared with the results of the parallel electric field and ethylene glycol/silicone oil experiments that are presented in this work.

Theoretical and numerical studies can be performed and compared with the results of the experimental work.

Different liquid pairs should be used in the experiments that are in the region of the diagram provided by Uguz *et al.* (2008) where both normal and parallel electric fields have destabilizing effects on the interface.

Different channel geometries can be designed and experiments can be carried out in the micro channels with smaller dimensions.

The parameters investigated in this work are the ratios of the flow rates and the viscosities, the effect of the width of the channel and the direction of the electric field. In addition to these, new parameters such as the conductivity ratio, can be studied.

The wavelength of the disturbance at the critical voltage may be measured and compared with the theory.

REFERENCES

- Abdella, K., and H. Rasmussen, 1997, “Electrohydrodynamic Instability of Two Superposed Fluids in Normal Electric Fields”, *Journal of Computational and Applied Mathematics*, Vol. 78, pp. 33-61.
- Anna, S.L., N. Bontoux, and H. A. Stone, 2003, “Formation of Dispersions Using Flow-focusing in Microchannels”, *Applied Physics Letters*, Vol. 82, No. 3, pp. 364-366.
- Barbier, V., H. Williaime, P. Tabeling, and F. Jousse, 2006, “Producing Droplets in Parallel Microfluidic Systems”, *Physical Review E: Statistical, Nonlinear, and Soft Matter Physics*, Vol. 74, No. 046306, pp. 1-4.
- Baroud, N. C., F. Gallaire, and R. Dangla, 2010, “Critical Review: Dynamics of Microfluidic Droplets”, *Lab on a Chip*, Vol 10, pp. 2032-2045.
- Baygents, J. C., and F. Baldessari, 1998, “Electrohydrodynamic Instability in a Thin Fluid Layer with an Electrical Conductivity Gradient”, *Physics of Fluids*, Vol. 10, pp. 301-311.
- Beulen, B., J. deJong, H. Reinten, M. van den Berg, H. Wisjshoff, and R. van Dongen, 2007, “Flows on the Nozzle Plate of an Inkjet Printhead”, *Experiments in Fluids*, Vol. 42, pp. 217-224.
- Bico, J., and D. Quéré, 2000, “Liquid Trains in a Tube”, *Europhysics Letters*, Vol. 51, pp. 546-550.
- Bransky, A., N. Korin, M. Khoury, and S. Levenberg, 2009, “A Microfluidic Droplet Generator Based on a Piezoelectric Actuator”, *Lab on a Chip*, Vol. 9, No. 4, pp. 516-520.
- Breslauer, D. N., P. J. Lee, and L. P. Lee, 2006, “Microfluidics-based Systems Biology”, *Molecular Biosystems*, Vol. 2, pp. 97-112.

- Cai, L., N. Friedman, and X. S. Xie, 2006, "Stochastic Protein Expression in Individual Cells at the Single Molecule Level", *Nature*, Vol. 440, pp. 358-362.
- Chen, C. T. and G. B. Lee, 2006, "Formation of Microdroplets in Liquids Utilizing Active Pneumatic Choppers on a Microfluidic Chip", *Journal of Microelectromechanical Systems*, Vol. 15, No. 6, pp. 1492-1498.
- Choi, Y. H., Y. S. Song, and D. H. Kim, 2011, "Microdroplets for the Study of Mass Transfer", In: Dr. Hironori Nakajima (Ed.), *Mass Transfer-Advanced Aspects*, pp. 805-824, Shangahi, China, InTech Press.
- Christopher, G. F., N. N., Noharuddin, J. A., Taylor, and S. L., Anna, 2008, "Experimental Observations of the Squeezing-to-Dripping Transition in T-shaped Microfluidic Junctions", *Physics Review Letters*, Vol. 78, No. 036317, pp. 1-12.
- Cramer, C., P. Fischer, E. J., Windhab, 2004, "Drop Formation in a Co-flowing Ambient Fluid", *Chemical Engineering Science*, Vol. 59, pp. 3045-3058.
- Craster, R. V., and O. K. Matar, 2005, "Electrically Induced Pattern Formation in Thin Leaky Dielectric Films", *Physics of Fluids*, Vol. 17, pp. 032104.1-032104.17
- Dang, F. Q., L. H. Zhang, M. Jabasini, N. Kaji, and Y. Baba, 2003, "Characterization of Electrophoretic Behavior of Sugar Isomers by Microchip Electrophoresis Coupled with Videomicroscopy", *Analytical Chemistry*, Vol. 75, pp. 2433-2439.
- de Mello, A. J., 2001, "DNA Amplification: Does Small Really Mean Efficient?", *Lab on a Chip*, Vol. 1, pp. 24-29.
- Dong, H. M., W. W. Carr, W., and J. F. Morris, 2006a, "An Experimental Study of Drop-on-Demand Drop Formation", *Physics of Fluids*, Vol. 18, pp. 072102.1-072102-16.

- Dong, H. M., W. W. Carr, and J. F. Morris, 2006b, "Visualization of Drop-on-Demand Inkjet: Drop Formation and Deposition", *Review of Scientific Instruments*, Vol. 77, pp. 085101.1-085101.8.
- El Moctar, A. O., N. Aubry and J. Batton, 2003, "Electro-hydrodynamic Micro-Fluidic Mixer", *Lab on a Chip*, Vol. 3, pp. 273-280.
- El-Sayed, M. F., G. M. Moatimid, and N. M. Hafez, 2011, "Electrohydrodynamic Instability of Two Superposed Viscous Dielectric Fluids Flowing Down an Inclined Plane with Thermal Conductivity Variation", *Progress in Applied Mathematics*, Vol. 2, No. 1, pp. 35-55.
- Ersoy, G., *Investigation of the Interfacial Instability between a Newtonian Fluid and a Polymeric Fluid under the Influence of an Electric Field for Microfluidics Applications*, MS. Thesis, Bogazici University, 2011.
- Evans, J., D. Liepmann, and A. P. Pisano, "Planar laminar mixer," Proceedings of the 1997 IEEE MEMS Workshop, Nagoya, Japan, pp. 96–101, 1997.
- Evensen, H. T., D. R. Meldrum, and D. L. Cunningham, 1998, "Automated Fluid Mixing in Glass Capillaries," *Review of Scientific Instruments*, Vol. 69, pp. 519–526.
- Fasina, O. O. and Colley, Z., 2008, "Viscosity and Specific Heat of Vegetable oils as a Function of Temperature: 35°C to 180°C", *International Journal of Food Properties*, Vol. 11, pp. 738–746.
- Fu, L.M., C. H. Lin, C.H., Chien, Y.S., 2004, "Microfluidic T-form Mixer Utilizing Switching Electroosmotic Flow", *Analytical Chemistry*, Vol. 76, pp. 5265–5272.
- Gamhire, P., and R. M. Thaokar, 2010, "Electrohydrodynamic Instabilities at Interfaces Subjected to Alternating Electric Field", *Physics of Fluids*, Vol. 22, pp. 064103.1-064103.16.

- Gañán-Calvo, A. M., 1998, “Generation of Steady Liquid Microthreads and Micro-sized Monodisperse Sprays in Gas Streams”, *Physical Review Letters*, Vol. 80, pp. 285-288.
- Garstecki, P., I. Gitlin, W. Di Luzio, G. M. Whitesides, E. Kumachecheva, and H. A. Stone, 2004, “Formation of Monodisperse Bubbles in a Microfluidic Flow-Focusing Device”, *Applied Physics Letters*, Vol. 85, No. 13, pp. 2649-2651.
- Garstecki, P., M. J. Fuerstman, H. A. Stone, and G. M. Whitesides, 2006, “Formation of Droplets and Bubbles in a Microfluidic T-junction – Scaling and Mechanism of Break-up”, *Lab on a Chip*, Vol. 6, pp. 437-446.
- Garstecki, P., M. J. Fuerstman, G. M. Whitesides, 2005, “Oscillations With Uniquely Long Periods in a Microfluidic Bubble Generator”, *Nature Physics*, Vol. 1, pp. 168-71.
- Gu, H., H. G. Michel, M. H. G. Duits, and F. Mugele, 2011, “Droplets Formation and Merging in Two-Phase Flow Microfluidics”, *International Journal of Molecular Science*, Vol. 12, No. 4, pp. 2572–2597.
- Gu, H., M. H. G. Duits, and F. Mugele, 2010, “A Hybrid Microfluidic Chip with Electrowetting Functionality Using Ultraviolet (UV) Curable Polymer”, *Lab on a Chip*, Vol. 10, pp. 1550-1556.
- Gu, H., F. Malloggi, A. G. Banpurkar, S. A. Vanapalli, and F. Mugele, 2008b, “Electrowetting - A Versatile Tool for Controlling Microdrop Generation”, *European Physical Journal E*, Vol. 26, No. 1-2, pp. 91-96.
- Gu, H., F. Malloggi, S. A. Vanopalli, F. Mugele, 2008a, “Electrowetting Enhanced Microfluidic Device for Drop Generation”, *Applied Physics Letters*, Vol. 93, pp. 183507.1-183507.3.
- Guillot, P. and A. Colin, 2005, “Stability of Parallel Flows in a Microchannel after a T Junction”, *Physical Review E*, Vol. 72, pp. 066301.1- 066301.4.

- Hatakeyema, T., D. L. L. Chen, R. F. Ismagilov, 2006, "Microgram-Scale Testing of Reaction Conditions in Solution Using Nanoliter Plugs in Microfluidics with Detection by MALDI-MS", *Journal of the American Chemical Society*, Vol. 128, pp. 2518-2519.
- He, P., H. Kim, D. W. Luo, M. Marquez, Z. D. Cheng, 2007, "Controlled Production of Emulsion Drops Using an Electric Field in a Flow Focusing Microfluidic Device", *Applied Physics Letters*, Vol. 91, No. 133106.
- Herminghaus, S., 1999, "Dynamical Instability of Thin Liquid Films between Conducting Media", *Physical Review Letters*, Vol. 83, No. 12, pp. 2359-2361.
- Hessel, V., H. Löwe, and F. Schönfeld, 2005, "Micromixers a Review on Passive and Active Mixing Principles", *Chemical Engineering Science*, Vol. 60, pp. 2479 – 2501.
- Hoburg, J. F., and J. R. Melcher, 1976, "Internal Electrohydrodynamic Instability and Mixing of Fluids with Orthogonal Field and Conductivity Gradients", *Journal of Fluid Mechanics*, Vol. 73, pp. 333-351.
- Hoburg, J. F., and J. R. Melcher, 1977, "Electrohydrodynamic Mixing and Instability Induced by Colinear Fields and Conductivity Gradients", *Physics of Fluids*, Vol. 20, pp. 903-911.
- Hosokawa, K., T. Fujii, and I. Endo, 1999, "Droplet-Based Nano/Picoliter Mixer Using Hydrophobic Microcapillary Vent", In: IEEE, *12th IEEE International Conference on Micro Electro Mechanical Systems*, Orlando, FL, USA, 1999, IEEE, pp. 388-393, USA.
- Hsiung, S. K., C. T. Chen, and G. B. Lee, 2006, "Microdroplet Formation Utilizing Microfluidic Flow-Focusing and Controllable Moving-Wall Chopping Techniques", *Journal of Micromechanics and Microengineering*, Vol.16, No. 11, pp. 2403-2410.

- Hsu, H. Y. and N. A., Patankar, 2014, “Electrokinetic Instability in the Sharp Interface Limit. Part II: The Perpendicular Electric Field Case”, *Fluid Dynamics*, eprint arXiv:1402.1776.
- Hua, J., B. Zhang and J. Lou, 2007, “Numerical Simulation of Microdroplet Formation in Coflowing Immiscible Liquids”, *AiChE Journal*, Vol. 53, No. 10, pp. 2354-2548.
- Husny, J. and J. J. Cooper-White, 2006, “The Effect of Elasticity on Drop Creation in T-Shaped Microchannels”, *Journal of Non-Newtonian Fluid Mechanics*, Vol. 137, pp. 121-136.
- Kath, G. S., and J. F. Hoburg, 1997, “Interfacial Electrohydrodynamic Instability in Normal Electric Field”, *Physics of Fluids*, Vol. 20, No. 6, pp. 912-917.
- Kim, S. J., Y. A. Song, P. L. Skipper, J. Han, 2006, “Electrohydrodynamics Generation and Delivery of Monodisperse Picoliter Droplets Using Poly(dimethylsiloxane) Microchip”, *Analytical Chemistry*, Vol. 78, No. 23, pp. 8011-8019.
- Köhler, J. M. and T. Henkel, 2005, “Chip Devices for Miniaturized Biotechnology”, *Applied Microbiology BioTechnology*, Vol. 69, pp. 113-125.
- Kumacheva, E. and P. Garstecki, 2011, *Physics of Microfluidic Emulsification, in Microfluidic Reactors for Polymer Particles*, John Wiley & Sons Ltd, Chichester, UK.
- Le, H. P., 1998, “Progress and Trends in Ink-Jet Printing Technology”, *Journal of Imaging Science and Technology*, Vol. 42, No. 1, p. 49.
- Lee, C. Y., Y. H. Lin, G. B. Lee, 2009, “A Droplet-Based Microfluidics System Capable of Droplet Formation and Manipulation”, *Microfluid Nanofluid*, Vol. 6, pp. 590-610.

- Li, F., O. Ozen, N. Aubry, D. T. Papageorgiou, and P. G. Petropoulos, 2007, "Linear Stability of a Two-Fluid Interface for Electrohydrodynamic Mixing in a Channel", *Journal of Fluid Mechanics*, Vol. 583, pp. 347–377.
- Li, H., T. N. Wong, N. T. Nguyen, 2012, "Instability of Two Pressure Driven Viscous Fluid Streams in a Microchannel Under a Normal Electric Field", *International Journal of Heat and Mass Transfer*, Vol. 55, No. 2324, pp. 6994-7004.
- Liau, A., R. Karnik, A. Majumdar, J. H. D. Cate, 2005, "Mixing in Biological Crowded Solutions in Miliseconds", *Analytical Chemistry*, Vol. 77, pp. 7618-7625.
- Lin, H., B. D. Storey, M. H. Oddy, C. H. Chen, and J.G. Santiago, 2004, "Instability of Electrokinetic Microchannel Flows with Conductivity Gradients", *Physics of Fluids*, Vol. 16, pp. 1922–1935.
- Liu, R. H., M. A. Stremmer, V. S. Kendra, G. O. Michael, J. G. Santiago, R. J. Adrian, H. Aref, and D. J. Beebe, 2000, "Passive Mixing in a Three-Dimensional Serpentine Microchannel", *Journal of Microelectromechanical Systems*, Vol. 9, No. 2.
- MEGlobal Group," MEGlobal-Ethylene Glycol Product Guide", 2008,
http://www.meglobal.biz/media/product_guides/MEGlobal_MEG.pdf, Accessed July 2014.
- Melcher, J. R., and C. V. Smith, 1969, "Electrohydrodynamic Charge Relaxation Overstability and Interfacial Perpendicular Field Instability", *Physics of Fluids*, Vol. 12, pp. 778-790.
- Melcher, J.R. and W. J. Schwarz, 1968, "Interfacial Relaxation Overstability in a Tangential Electric-Field", *Physics of Fluids*, Vol. 11, pp. 2604-2616.
- Moroney, R. M., R. M. White, and R. T. Howe, 1991,"Ultrasonically Induced Microtransport", In: IEEE, *Proceedings: An Investigation of Micro Structures, Sensors, Actuators, Machines and Robots*, Nara, 1991, IEEE, pp. 277–282, 1991.

- Murshed, S. M. S., N. T. Nguyen, and S. H. Tan, 2008a, "Temperature Dependence of Interfacial Properties and Viscosity of Nanofluids for Droplet-Based Microfluidics", *Journal of Physics D: Applied Physics*, Vol. 41, No. 8.
- Murshed, S. M. S., N. T. Nguyen, S. H. Tan, T. N. Wong, and Yobas, L., 2008b, "Thermally Controlled Droplet Formation in Flow Focusing Geometry: Formation Regimes and Effect of Nanoparticle Suspension", *Journal of Physics D: Applied Physics* Vol. 41, No. 16.
- Nguyen, N. T., T. H. Ting, Y. F. Yap, T. N. Wang, J. C. K. Chai, W. L. Ong, J. Zhou, S. H. Tang, and L. Yobaş, 2007, "Thermally Mediated Droplet Formation in Microchannels", *Applied Physics Letters*, Vol. 91, pp. 084102.1-084102.3.
- Nie, H.Z., M. S. Seo, Q. S. Xu, P. C. Lewis, M. Mok, M. E. Kumacheva, G. M. Whitesides, P. Garstecki, and H. A. Stone, 2008, "Emulsification in Microfluidic Flow-Focusing Device: Effect of Properties of Liquids", *Microfluid. Nanofluid.*, Vol. 5, pp. 585-594.
- Nsisako, T., T. Torri, T. Higuchi, 2002, "Droplet Formation in a Microchannel Network", *Lab on a Chip*, Vol. 2, pp. 24-26.
- Nunes, J. K., S. S. H. Tsai, J. Wan, and H. A. Stone, 2012, "Dripping and Jetting in Micro Fluidic Multiphase Flows Applied to Particle and Fibre Synthesis", *Journal of Applied Physics D: Applied Physics*, Vol. 46, No. 11, pp. 1-20.
- Nurocak, A., *Effect of the Direction of the Electric Field on the Interfacial Instability between a Newtonian Fluid and Viscoelastic Polymer*, MS. Thesis, Bogazici University, 2012.
- Oddy, M. H., J. G., Santiago and J. C. Mikkelsen, 2001, "Electrokinetic Instability Micromixing", *Analytical Chemistry*, Vol. 73, pp. 5822-5832.

- Okushima, S., T. Nsisako, and T. Torri, 2004, "Controlled Production of Monodisperse Double Emulsions by Two-Steps Droplet Breakup in Microfluidic Devices", *Langmuir*, Vol. 20, No. 23, pp. 9905-9908.
- Onal, Y., M. Lucas, P. Claus, 2005, "Application of Capillary Microreactor for Selective Hydrogenation of α,β -Unsaturated Aldehydes in Aqueous Multiphase Catalysis", *Chemical Engineering Technology*, Vol. 28, p. 972.
- Ozen O., N. Aubry, D. T. Papageorgiou, and P. G. Petropoulos, 2006a, "Monodisperse drop Formation in Square Microchannels", *Physics Review Letters*, Vol 96, No. 14.
- Ozen,O., D. T. Papageorgiou, and P. G. Petropoulos, 2006b, "Nonlinear Stability of a Charged Electrified Viscous Liquid Sheet under the Action of a Horizontal Electric Field", *Physics of Fluids*, Vol. 18, No. 042102.
- Ozen, O., N. Aubry, D. Papageorgiou, and P. Petropoulos, 2006c, "Electrohydrodynamic Linear Stability of Two Immiscible Fluid in Channel Flow", *Electrochimica Acta*, Vol. 51, pp. 5316-5323.
- Paranjep, G. R., and P. Y. Deshpande, 1935, "Dielectric Properties of Some Vegetable Oils", *Proceedings of the Indian Academy of Sciences*, Vol. 1, No. 12, pp 880-886.
- Patankar,N. A.,2011,"Electrokinetic Instability: The Sharp Interface Limit", *Physics of Fluids*, Vol. 23, pp. 14101-14115.
- Pease III, L. F., and W. B. Russel, 2002, "Linear Stability Analysis of Thin Leaky Dielectric Films Subjected to Electric Fields", *Journal of Non-Newtonian Fluid Mechanics*, Vol. 102, pp. 233-250.
- Poe, S. L., M. A. Cummings, M. P. Haal, and D. T. McQuade, 2006, "Solving the Clogging Problem: Precipitate-Forming Reactions in Flow", *Angewandte Chemie International Edition*, Vol. 45, pp. 1544–1548.

- Posner, J.D. and J. G. Santiago., 2006, “Convective Instability of Electrokinetic Flows in a Cross-Shaped Microchannel”, *Journal of Fluid Mechanics*, Vol. 555, pp. 1-42.
- Rayleigh, L., 1882, “On the Equilibrium of Liquid Conducting Masses Charged with Electricity”, *Philosophical Magazine*, Vol. 14, pp. 184-186.
- Roach, S. L., H. Song, and R. F. Ismagilov, 2005, “Controlling Nonspecific Protein Adsorption in a Plug-Based Microfluidic System by Controlling Interfacial Chemistry Using Fluorous-Phase Surfactants”, *Analytical Chemistry*, Vol. 77, No. 3, pp. 785-796.
- Roberts, S. A. and S. Kumar, 2009, “AC Electrohydrodynamic Instabilities in Thin Liquid Films”, *Journal of Fluid Mechanics*, Vol. 631, pp. 255-279.
- Romero, P. A. and A. R., Abate, 2012, “Flow Focusing Geometry Generates Droplets Through a Plug and Squeeze Mechanism”, *Lab on a Chip*, Vol. 12, pp. 5130-5132.
- Saied, M. A., S. H. Mansour, M. Eweis, M. Z. El-Sabee, A. L. G. Saad, K. N., and K. N. A. Nour, 2008, “Some Biophysical Properties of Castor Oil Esterified with Some Acid Anhydrides”, *European Journal of Lipid Science and Technology*, Vol. 110, pp. 926-934.
- Santiago, J. G., H. Lin, H. Lele and C. H. Chen, 2005, “Convective and Absolute Electrokinetic Instability with Conductivity Gradients”, *Journal of Fluid Mechanics*, Vol. 524, pp. 263-303.
- Sassa, F., J. Fukuda, H. Suzuki, 2008, “Microprocessing of Liquid Plugs for Bio/Chemical Analyses”, *Analytical Chemistry*, Vol. 80, pp. 6206-6613.
- Saville, D. A., 1997, “Electrohydrodynamics: The Taylor-Melcher Leaky Dielectric Model”, *Annual Review of Fluid Mechanics*, Vol.29, pp. 27-64.

- Schäffer, E., T. Thurn-Albrecht, T. P. Russell, and U. Steiner, 2001, “Electrohydrodynamic Instabilities in Polymer Films”, *Europhysics Letters*, Vol. 53, No. 4, pp. 518–523.
- Schwesinger, N., T. Frank, and H. Wurmus, 1996, “A Modular Microfluid System with an Integrated Micromixer”, *Journal of Micromechics and Microengineering*, Vol. 6, No. 1, pp. 99-102.
- Shankar, V., and A. Sharma, 2004, “Instability of the Interface between Thin Fluid Films Subjected to Electric Field”, *Journal of Colloid Interface Science*, Vol. 274, pp. 294-308.
- Shin Etsu Company, “Silicone Fluid Technical Data Sheet”, 2004,
http://www.silicone.jp/e/catalog/pdf/kf96_e.pdf, Accessed July 2014.
- Shri Ram Oil & Chemical Industries, “Castor Oil Manufacturing”, 2012,
<http://www.shriramcastor.com/products.html>, Accessed July 2014.
- Song, H., D. L. Chen, and R. F. Ismagilov, 2006, “Reactions in Droplets in Microfluidic Channel”, *Angewandte Chemie International Edition*, Vol. 45, No. 44, pp. 7336-7356.
- Song, H. and R. F. Ismagilov, 2003, “Milisecond Kinetics on a Microfluidic Chip Using Nanoliters of Reagents”, *Journal of the American Chemical Society*, Vol. 125, pp. 14613-14619.
- Stone, H.A., A. D. Stroock, and A. Ajdari, 2004, “Engineering Flows in Small Devices: Microfluidics toward a Lab-on-a-Chip”, *Annual Review of Fluid Mechanics*, Vol. 36, pp. 381–411,
- Stroock A. D., S. K. Dertinger, A. Ajdari, I. Mezic, H. A. Stone HA, and G. M. Whitesides, 2002, “Chaotic Mixer for Microchannels”, *Science*, Vol. 295, No. 5555, pp. 647-51.

- Supeene, G., C. R. Koch, and S. Bhattacharjee, “Numerically Modelled Dynamic Response of Perfect and Leaky Dielectric Droplet in an Electric Field”, In: Proceeding ICMENS, *International Conference on MEMS, NANO and Smart Systems*, Banff, Alberta, Canada, 24-27 July 2005, IEEE Computer Society Washington, DC, USA, 2005.
- Tan S. H., N. T. Nguyen, L. Yobaş, and T. G. Kang, 2010, “Formation and Manipulation of Ferrofluid Droplets at a Microfluidic T-Junction”, *Journal of Micromechanics and Microengineering*, Vol. 20, No. 4, No. 045004.
- Tan, S. H., S. M. Murshed, N. T. Nguyen, T. N. Wong, and L. Yobas, 2008, “Thermally Controlled Droplet Formation in Flow Focusing Geometry: Formation Regimes and Effect of Nanoparticle Suspension”, *Journal of Physics D: Applied Physics*, Vol. 41, No. 165501.
- Tan, Y. C. and Lee, A. P., 2005, “Microfluidic Separation of Satellite Droplets as the Basis of a Monodispersed Micron and Submicron Emulsification System”, *Lab on a Chip*, Vol. 6, pp. 1178-1183.
- Taylor, G. I. and D. McEwan, 1965, “The Stability of a Horizontal Fluid Interface in a Vertical Electric Field”, *Journal of Fluid Mechanics*, Vol. 1965, No. 1, pp. 1-15.
- Taylor, G. I., and J. R. Melcher, 1969, “Electrohydrodynamics: A Review of Role of Interfacial Shear Stresses”, *Annual Review of Fluid Mechanics*, Vol. 1, pp. 111-146.
- Teh, S.-Y., R. Lin, L.-H. Hung, A. P. Lee, 2008, “Droplet Microfluidics”, *Lab on a Chip*, Vol. 8, pp. 198–220.
- Thaokar, R. M., and V. Kumaran, 2005, “Electrohydrodynamic Instability of the Interface Between Two Fluids Confined in a Channel”, *Physics of Fluids*, Vol. 17, No. 084104.

- Thorsen, T., R. W. Roberts, F. H. Arnold, S. R. Quake, 2001, "Dynamic Pattern Formation in a Viscle-Generating Microfluidic Device", *Physical Review Letters*, Vol. 86, No. 18, pp. 4163-4166.
- Tice, J. D., A. D. Lyon, R. F. Ismagilov, 2004, "Effects of Viscosity on Droplet Formation and Mixing in Microfluidic Channels", *Analytica Chimica Acta*, Vol. 507, pp. 73-77.
- Tsai, H. H., Wang, J. J., Su, Y. C., 2012, "Electric Field Induced Tip Streaming for Sub-femtoliter Droplet Formation", *16th International Conference on Miniaturized Systems for Chemistry and Life Sciences*, Okinawa, Japan, October 28th- November 1st 2012, MicroTAS, Japan, 2012.
- Uguz, A. K., and N. Aubry, 2008, "Quantifying the Linear Stability of a Flowing Electrified Two-Fluid Layer in a Channel for Fast Electric Times for Normal and Parallel Electric Fields", *Physics of Fluids*, Vol. 20, No. 092103.
- Uguz, A. K., O. Ozen, and N. Aubry, 2008, "Electric Field Effect on a Two-Fluid Interface Instability in Channel Flow for Fast Electric Times", *Physics of Fluids*, Vol. 20, No. 031702.
- Umbanhowar, P.B., V. Prasad, D. A. Weitz, 2000, "Monodisperse Emulsion Generation via Drop Break Off in a Co-flowing Stream", *Langmuir*, Vol. 16, No. 2, pp. 347-351.
- Utada, A. S., A. F., Nieves, H. A., Stone, and D. A., Weitz, 2007, "Dripping to Jetting Transitions in Coflowing Liquid Streams", *Physical Review Letters*, Vol. 99, No. 094502.
- Utada, A. S., E. Lorenceau, D. R. Link, P. D. Kaplan, H. A. Stone, and D. A. Weitz, 2005, "Monodisperse Double Emulsions Generated from a Microcapillary Device", *Science*, Vol. 308, No. 5721, pp. 537-541.
- Vogelstein, B., K. W. Kinzler, K., 1999, "Digital PCR", *Proceedings of the National Academy of Sciences. USA*, Vol. 96, pp. 358-362.

- West, J., B. Karamata, B. Lillis, J. P. Gleeson, J. Alderman, J. K. Collins, W. Lane, A. Matthewson, and H. Berney, 2002, "Application of Magnetohydrodynamic Actuation to Continuous Flow Chemistry", *Lab on a Chip*, Vol. 2, pp. 224–230.
- Whitesides, G.M., 2006, "The Origins and the Future of Microfluidics", *Nature*, Vol. 442, pp. 368–373.
- Willaime, H., V. Barbier, L. Kloul, S. Maine, and P. Tabeling, 2006, "Arnold Tongues in a Microfluidic Drop Emitter", *Physical Review Letters*, Vol. 96, No. 054501.
- Wu, L., and S. Y. Chou, 2005, "Electrohydrodynamic Instability of a Thin Film of Viscoelastic Polymer Underneath a Lithographically Manufactured Mask", *Journal of Non-Newtonian Fluid Mechanics*, Vol. 125, pp. 91–99.
- Xu, J., G. S. Luo, G. G. Chen, and J. D. Wang, 2005, "Experimental and Theoretical Approaches on Droplet Formation from a Micrometer Screen Hole", *Journal of Membrane Science*, Vol. 266, pp. 121–131.
- Xu, J. H., S. W. Li, J. Tan, Y. J. Wang, and G. S. Luo, 2006a, "Controlled Preparation of Monodisperse O/W and W/O Emulsions in the Same Microfluidic Device", *Langmuir*, Vol. 22, No. 19, pp. 7943–7946.
- Xu, J. H., S. W. Li, J. Tan, Y. J. Wang, and G. S. Luo, 2006b, "Preparation of Highly Monodisperse Droplet in a T-Junction Microfluidic Device", *AIChE Journal*, Vol. 52, No. 9, pp. 3005–3010.
- Yang, J.T., K. J. Haung, K. Y. Tung, I. C. Hu, P. C. Lyu, 2007, "A Chaotic Micromixer Modulated by Constructive Vortex Agitation", *Journal of Micromechanics and Microengineering*, Vol. 17, pp. 2084–2092.
- Zhang, H. B., M. J. Edirisinghe, S. N. Jayasinghe, 2006, "Flow Behaviour of Dielectric Liquids in an Electric Field", *Journal of Fluid Mechanics*, Vol. 558, pp 103–111

Zheng, B., J. D. Tice, R. F. Ismagilov, 2004, "Formation of Droplets of Alternating Composition in Microfluidic Channels and Applications to Indexing of Concentrations in Droplet-Based Assays", *Analytical Chemistry*, Vol. 76, pp. 4977-4982.

Zhou, C, P. Yue, J. J. Feng, 2006, "Formation of Simple and Compound Drops in Microfluidic Devices", *Physics of Fluids*, Vol. 18, No. 092105, pp. 1-14.