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Experimental investigation of electrohydrodynamic instabilities in micro channels

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Abstract. An electric field is applied to destabilize the interface between two Newtonian and immiscible liquids flowing in a rectangular micro channel. 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 between these liquids. The two liquids used in the experiments are a combination of ethylene glycol, different viscosity silicone oils, castor oil, and olive oil. The onset of electrohydrodynamic instability is investigated for various parameters, including the ratios of the flow rates, and viscosities of the liquids, the width of the micro channel, and the direction of the applied electric field. The order of the voltage applied to destabilize the interface is in the range 95 and 1190 V. The results of the experiments show that an increase in the viscosity ratio and the flow rate ratio of silicone oil to ethylene glycol have a stabilizing effect. It is also found that the important parameter to determine the critical voltage is the flow rate ratio, not the individual flow rates of the liquids. Also, as the width of the micro channel increases, the critical voltage increases. Lastly, for the liquid combinations used in the experiments, the interface could not be destabilized under the influence of a parallel electric field.

1 Introduction

In recent years, there has been a growing interest in microfluidics due its applications in many areas, including life sciences, reaction engineering, fuel cells, and many others. The idea is to use less volume and have more control over the system. However, in a micro channel as the characteristic length diminishes to microns, Reynolds number is usually very small; hence, the flow is laminar. Consequently, one major research area that has attracted many researchers is to find a way to effectively mix two miscible fluids that flow in a micro channel [1–4]. A second research area is to generate micro droplets of controllable size and size distribution in micro channels from immiscible liquids. These droplets, which are of pico liter or even of less volume may serve as reaction vessels and they are generated from two or multi phase flows in micro

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channels [5–9]. The reader is referred to Teh et al. (2008) and Seemann et al. (2012) for a detailed review of droplet microfluidics [10,11]. Most of the methods to obtain an efficient mixing in micro channels for miscible liquids and to generate droplets from immiscible liquids in micro channels involve changing the straight channel geometry. However, it is also possible to obtain an efficient mixing in a micro channel by applying an electric field either normal [2] or parallel [3] to the flat interface between the two liquids. In both cases, the electric field needed for mixing is on the order of 10^5 V/m even though the absolute voltage for the normal field is much smaller as the distance between the electrodes is very short. It is also possible to generate droplets between two immiscible liquids flowing in a channel when an electric field is applied normal to the flat interface [9]. Ozen et al. (2006) used constant volumetric flow rates of glycerine and corn oil in a micro channel and applied a DC electric field [9]. At some critical voltage the interface between the two liquids started deflecting, flapping one of the walls, and capturing glycerine slugs, which became spherical due to the interfacial tension after they left the electric field region. Ozen et al. (2006) were able to obtain mono dispersed micro droplets. They observed that the size of the droplets decreased with increasing voltage. The deflection of the interface between the two liquids is due to electrohydrodynamic (EHD) instability [12–19]. The physics of the instability for perfect dielectric or leaky dielectric liquids is due to the discontinuities in the mean electric potentials at the interface [15] and the modeling of leaky dielectric fluids is developed by Taylor and Melcher [21,22].

Several approaches, including linear stability analysis, lubrication theory, weakly nonlinear analysis and fully nonlinear analysis are used in the literature to understand the electrohydrodynamic instabilities. There are several works that assume fast electric charge relaxation as it brings an important simplification [15–18,23]. For fast charge relaxation times, Uguz et al. (2008) showed that it is possible to find a relationship between the conductivity and permittivity ratios of the fluids without even solving the momentum equations to determine whether the electric field has a stabilizing or destabilizing effect [18]. In fact, once the electric field has a stabilizing effect it turns out that the interface is always stable [19].

There are fewer experimental studies in the literature. Most of the experimental studies are attributed to non-Newtonian fluids to better understand the lithographically-induced self-assembly (LISA) process [24,25] along with some numerical work [26,27]. Gambhire and Thaokar (2012) studied the stability of the interface between a thin silicone oil layer and air system, both theoretically and experimentally for both an AC and a DC field [29]. There are also some experiments especially from Santiago's group [3,28] about the effect of the parallel electric field on the effective mixing of two miscible liquids. They used a high conductive liquid and a low conductive liquid in their experiments and assumed a conductivity gradient orthogonal to the direction of the flow. Aubry's group was also interested in the effective mixing of miscible liquids [2]. However, they used an electric field normal to the flat interface under either AC or DC field.

To the best of our knowledge, there are only two papers where the aim is to determine the critical voltage at which the interface becomes unstable for two immiscible liquids under either normal [30] or parallel [31] electric field. Li et al. (2012) [30,31] experimentally and analytically investigated the interface between two immiscible liquids, one is a nonconducting liquid (silicone oil) and the other is a conducting liquid ($NaHCO_3$). They visually observed that normal and parallel electric fields can have a destabilizing effect. They also found out that an increase in the viscosity (of the conducting liquid) and the total flow rate have a stabilizing effect on the interface of these two immiscible liquids. Also, they showed that [31] there is around 30% difference between the experimental results and the analytical findings of the study due to the complications in determining the critical point.

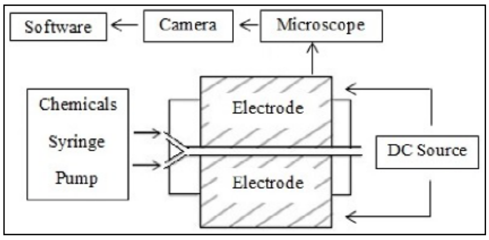


Fig. 1. Sketch of the experimental setup for normal electric field applications.

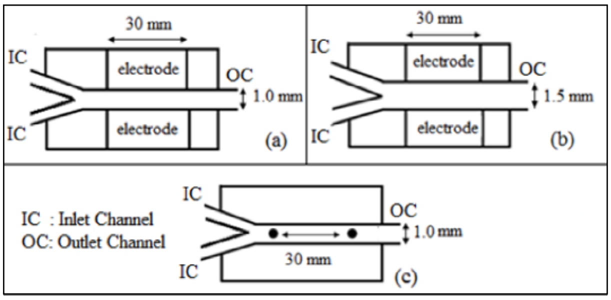


Fig. 2. Sketch of the micro channels with electrodes to apply an electric field (a) normal to the interface, width is 1.0 mm (b) normal to the interface, width is 1.5 mm of width (c) parallel to the interface, distance is 30 mm.

This paper aims to determine the experimental onset of instability for two immiscible liquids flowing in a micro channel subject to a DC electric field applied either parallel or normal to the flat interface between these liquids. The previous studies, as discussed above are either for miscible liquids [2,3,28] where the aim is to obtain an efficient mixing or for immiscible liquids where one of the liquids is conductive (electrolyte) and the other one is dielectric [30,31] whereas in our case there is no electrolyte, which suggests a different theory [22,30,31]. On the other hand, Ozen et al. (2006) used chemicals with electrical properties that are similar to those used in our study [9]. However, the focus of that paper is to determine the relationship between the droplet size and the applied voltage for corn oil and glycerine flowing in a micro channel and not to determine the critical voltage at which the interface becomes unstable [9].

Section 2 presents the experimental setup and the procedure, Sect. 3 includes the results and discussions, and Sect. 4 is the conclusion.

2 Experimental setup and procedure

The experimental setup shown as a sketch in Fig. 1 consists of a micro channel, a dual syringe pump (Harvard Apparatus), a microscope (Nikon Eclipse), a high speed camera (Fastec InLine) connected to a computer with an imaging software (Fastec Imagining Co.), and a DC power source (Glassman High Voltage Inc) with a maximum of 6, 000 V supply. 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 sketch of the micro channels used in the experiments is shown in Fig. 2 and they are of Y-shape with two stainless steel inlets of 1.5 mm outside diameter. All of the micro channels are made of PMMA and they are 50 mm long and 200 μm deep.

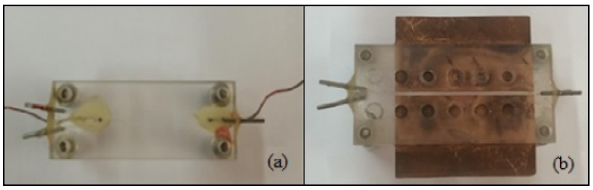


Fig. 3. Micro channels with electrodes to apply (a) parallel or (b) normal; electric field to the flat interface between the immiscible liquids.

Table 1. Physical properties of the chemicals used in the experiments.

Chemicals	Density (kg/m^3)	Kinematic viscosity (cSt)	Conductivity (S/m)	Relative permittivity
Ethylene glycol	1110	15	1.1E-04	37.70
Silicone oil 5	915	5	1.0E-13	2.57
Silicone oil 10	930	10	1.0E-13	2.65
Silicone oil 20	950	20	1.0E-13	2.69
Silicone oil 50	960	50	1.0E-13	2.75
Silicone oil 100	965	100	1.0E-13	2.79
Castor oil	961	1025	5.0E-12	4.78
Olive oil	911	92	3.0E-11	3.35

For the normal electric field application, two copper sheets each having a length of 30 mm, a width of 15 mm and a thickness of 200 μm are placed facing one another as shown in Fig. 2a and Fig. 2b. The two copper sheets are placed between the top and the bottom PMMA plates, so that the depth of the micro channel is 200 μm for the normal electric field applications. Two micro channels of 1.0 mm (Fig. 2a) and 1.5 mm (Fig. 2b) 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. 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 Fig. 2c. The distance between the electrodes is 30 mm. The width and the depth of the micro channel for the parallel electric application are 1.0 mm and 200 μm, respectively. Figure 3 shows the actual micro channels of width 1.0 mm with copper electrodes that allow either parallel (Fig. 3a) or normal (Fig. 3b) electric field applications.

A pressure gradient is applied to the liquids by the syringe pump. The two immiscible liquids flow into the micro channel through their own inlets until a flat interface is formed between them. Then, the DC power source is turned on and a potential difference is applied either parallel or normal to the flat interface. The applied voltage is increased by increments of 10 V. At each increment the flatness of the interface is checked. The critical voltage is determined when a deflection of the interface is observed. Each experiment is repeated at an average of 5 times, and the average of the results is reported along with the standard deviation as the final critical voltage. As the applied potential difference is decreased below the critical voltage, the deflected interface recovers its flat shape.

In the experiments; ethylene glycol, silicone oils of various viscosities, castor oil and olive oil are used as the liquids. The physical properties of the chemicals are listed in Table 1. The surface tension between ethylene glycol and silicone oil is reported to be 18 mN/m [32].

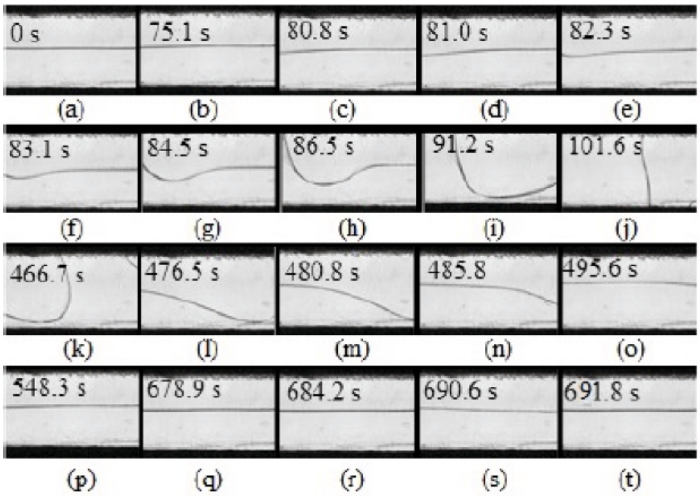


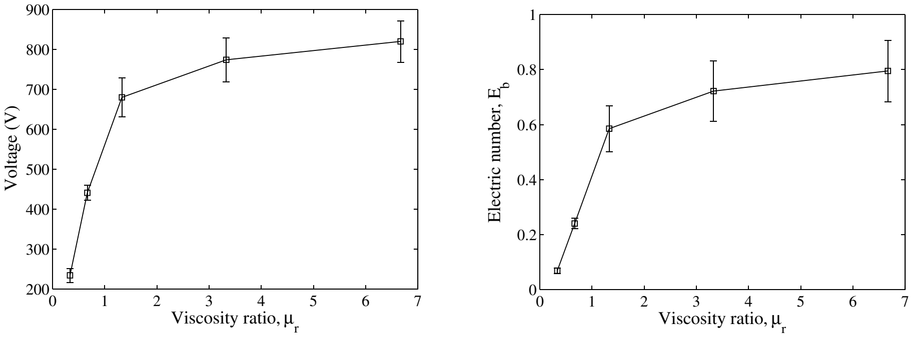
Fig. 4. The evolution of the interfacial instability between ethylene glycol and 50 cSt silicone oil flowing at 10 μL/min in a micro channel of 1.0 mm width. The critical voltage of 880 V is reached in (b). The rupture occurred in (h). The electric field is turned off in (o) and the flat interface is recovered in (t).

3 Results and discussion

The interface between two immiscible liquids with different electrical properties can be destabilized due to the electrical charge deposition at the interface when the liquids are subject to an electric field. In this work, the effects of four different parameters are studied, namely the viscosity and the 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.

When an electric field is applied to the flat interface between two immiscible liquids in a micro channel, the interface starts deflecting at the critical voltage of the system depending on the permittivities and conductivities of the liquids. The direction of the electric field may be normal or parallel to the flat interface between the liquids. Figure 4 shows the deflection of the interface as a function of time between ethylene glycol and 50 cSt silicone oil, both flowing at 10 μL/min side by side in the micro channel under a normal electric field. Here, the top liquid is ethylene glycol and the bottom one is silicone oil, 50 cSt. Initially, at time $t = 0$ (Fig. 4a) the interface is flat. The intensity of the electric field, normal to the flat interface is kept increasing and at each voltage the interface is observed for any deflection. At $t = 75.1$ s (Fig. 4b) the applied voltage reaches the critical voltage of the system, 880 V and the interface starts deflecting. At that point, the voltage is not increased anymore and the fate of the interface is recorded as a function of time, not position (Fig. 4b through Fig. 4o). For some experiments as in this one, the amplitude of the interface deflection may increase and the interface may flap one of the walls as observed in Fig. 4h. This may cause the rupture of the interface (Fig. 4j). If the applied voltage is decreased slowly (Fig. 4o), it may be possible to recover the flat interface as seen in Fig. 4t. The video of the experiment shown in Fig. 4 can be found in the supplementary material section (see VideoES50).

The effect of the viscosity ratio, μ_r is investigated for the cases of normal and parallel applied electric fields. The viscosity ratio is altered from 0.033 to 6.667 for the



(a) (b)

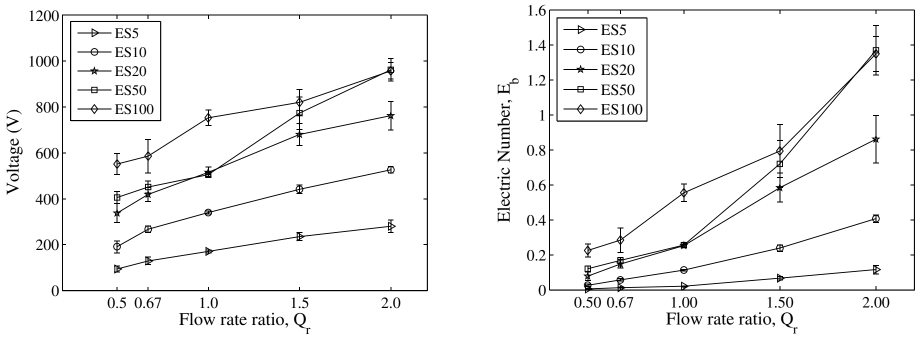
Fig. 5. The effect of the viscosity ratio on (a) the voltage and (b) the electric number, E_b; in the presence of a normal electric field with a channel width of 1.0 mm.

systems of ethylene glycol flowing at 30 μL/min and silicone oil of different viscosities flowing at 20 μL/min. Figure 5a shows the voltage (V) needed to destabilize ethylene glycol- silicone oil systems with different viscosity ratios in the presence of a normal electric field in a micro channel with a width of 1 mm. It should be noted that each experiment is run at an average of 5 times and the critical voltages are reported as the average of these runs along with the standard deviations. Experimental studies show that an increase in the viscosity ratio has a stabilizing effect on the interface in the presence of a normal electric field. Hence a system with higher viscosity ratio requires higher potential difference for destabilization. This result is also expected theoretically [14,16] due to the viscous dissipation of the liquids. There are two trends in Fig. 5. First, as the viscosity ratio increases the critical voltage increases. Then, as the viscosity ratio keeps increasing, there is a plateau for the critical voltage. The stabilizing effect of the viscosity ratio is due to the viscous dissipation. However, at some point the viscosity is so high that it is not changing the destabilizing effect of the electrical stresses. This plateau is also observed numerically in [14] in their Figs. 3b and 7. Similarly, even for one conducting and one dielectric liquid Li et al. [30,31] observed that an increase in the viscosity of conducting liquid had a stabilizing effect. It is also possible to present our results in dimensionless form as in Fig. 5b. Here, a dimensionless electric number, E_b is plotted against the viscosity ratio. The electric number is defined as [18]

$$E_b = \left[\varepsilon_0 V^2\right] / [\mu^{(2)} U h^{(2)}],$$

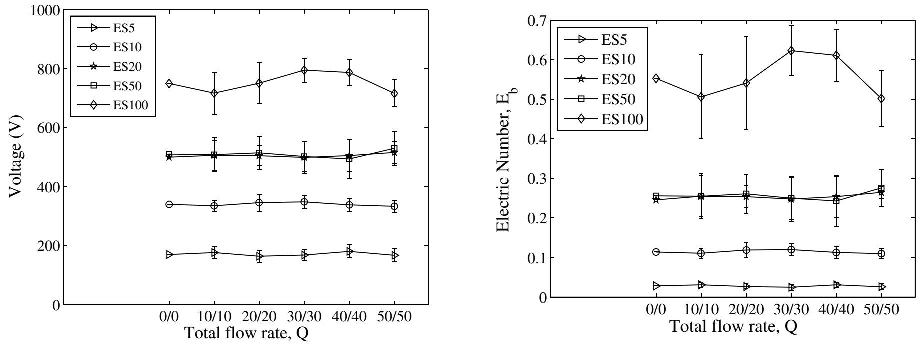
where ε_0, U, and V are the vacuum permittivity, the characteristic velocity defined as $\gamma/\mu^{(2)}$, and the applied voltage, respectively. Here, γ is the interfacial tension between the liquids. Also $h^{(1)}$ and $h^{(2)}$ are the thicknesses occupied by the first and second liquids in the micro channel, respectively. The sum of $h^{(1)}$ and $h^{(2)}$ gives the width of the micro channel.

Another important parameter is the effect of the flow rate ratio, Q_r where Q_r is the ratio of the flow rate of the first liquid, $Q^{(1)}$ to the flow rate of the second liquid, $Q^{(2)}$. This parameter is studied by arranging the flow rates of silicone oil, $Q^{(1)}$ and ethylene glycol, $Q^{(2)}$ in such a way the ratios of the flow rates are 0.50, 0.67, 1.00, 1.50 and 2.00. These ratios are obtained using $Q^{(1)}/Q^{(2)}$ as $20/40, 20/30$, average of $(10/10, 20/20, 30/30, 40/40, 50/50)$ experiments, 30/20, and 40/20. Figure 6 shows the effect of the flow rate ratio on the stability of silicone oil and ethylene glycol systems in terms of voltage (Fig. 6a) and electric number, E_b (Fig. 6b) for the normal



(a) (b)

Fig. 6. The effect of the flow rate ratio on (a) the voltage and (b) the electric number, E_b; in the presence of a normal electric field in a micro channel of width 1.0 mm.

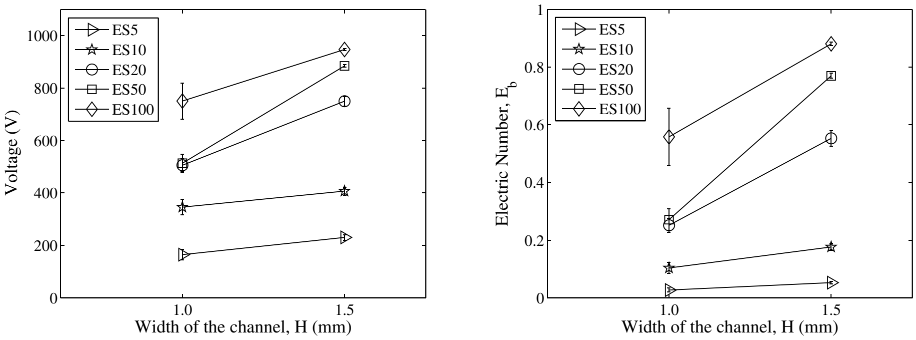


(a) (b)

Fig. 7. The effect of individual flow rates of silicone oil and ethylene glycol such that the ratio of flow rates is unity in all cases (a) on the voltage and (b) on the electric number, E_b; in the presence of a normal electric field.

electric field application in a micro channel of width 1.0 mm. In the legends of Figs. 6 to 8, ES5, ES10, ES20, ES50 and ES100 indicate the liquid combinations of ethylene glycol with silicone oils of 5 cSt, 10 cSt, 20 cSt, 50 cSt and 100 cSt. As seen from Fig. 6a, the results show that the critical voltage needed to destabilize a system is directly proportional with the flow rate ratio. An increase in the flow rate of silicone oil induces an increase in the flow rate ratio, which has a stabilizing effect.

We also investigate a special case of the effect of the flow rate ratio where the total flow rate Q is altered in such a way that the ratios of the flow rates are kept constant at unity. The total flow rate used in these experiments is varied from 10/10 μL/min to 50/50 μL/min. Figure 7a shows the potential difference needed to destabilize ethylene glycol-silicone oil interface for five types of silicone oils, in the presence of a normal electric field when both liquids flow at the same rate. As shown in Fig. 7a, the important parameter to determine the critical voltage is not the individual flow rates, but it is their ratio. Furthermore, the same effect is investigated for no flow case, i.e. 0/0 case as seen in the figure. For this purpose the flow is stopped and the quiescent state is reached. When the interface is stable and the thicknesses of the liquid layers are equal to each other, a normal electric field is applied and its magnitude is increased slowly. This system is also destabilized at the same potential difference



(a) (b)

Fig. 8. The effect of the width of the micro channel for silicone oil-ethylene glycol pairs on (a) the critical voltage and (b) the electric number, E_b; in the presence of a normal electric field.

(Figs. 7a and b). Even though when the liquids flow in the channel, a destabilized interface can regain its flat shape after the voltage is turned off, for the no-flow case, the destabilized interface cannot regain its flat shape even after turning off the voltage.

Next, the effect of the channel width is investigated. The width of a micro channel actually indicates the distance between the electrodes. To understand the effect of the width of the micro channel on the stability of the interface, two micro channels of 1.0 mm (Fig. 3b) and 1.5 mm width are used and their effects are compared in the presence of a normal electric field. For each viscosity ratio, as depicted in Fig. 8a, the critical potential difference (V) needed to destabilize the interface between ethylene glycol and each type of silicone oil flowing at $10\,\mu\mathrm{L/min}$ increases for the larger channel width. Like the potential difference, the dimensionless electric number, E_b of the system also increases for increasing channel width (Fig. 8b).

Finally, the effect of the direction of the electric field is investigated. For this purpose, the electric field is applied either normal or parallel to the flat interface between the two immiscible liquids where the width of the micro channel is fixed at 1.0 mm. The normal electric field results are already presented. The parallel electric field is applied to ethylene glycol-silicone oils, castor oil-silicone oils and olive oil-silicone oils systems at various flow rates, i.e. thickness ratios of liquids. Silicone oils are of 5, 10, 20, 50, and 100 cSt viscosities. None of these systems were destabilized up to a maximum of $6,000$ V that was applied. The results are compared with an analytical work [18] where two leaky dielectric liquids are assumed to flow in a channel under the effect of either normal or parallel electric field. According to Uguz et al. (2008) [18] the effect of the electric field can be deduced from conductivity ratios versus permittivity ratios of the liquids. The liquid pairs used in our experiments fall in a region where the parallel electric field has a stabilizing effect, i.e., the system is always stable independent of the applied voltage. It should be noted that in that region, the normal electric field has a destabilizing effect. Therefore, our experiments match the results of [18]. The results in [18] are valid only for leaky dielectric liquids and fast relaxation times. However, when one conducting and one dielectric liquids are subject to a parallel electric field, it is shown that the interface might become unstable [31].

4 Conclusion

Electrohydrodynamic instability of the flat interface between two immiscible liquids pumped into a micro channel via a syringe pump is studied experimentally in the presence of an electric field, which is either normal or parallel to that interface. The parameters that are investigated are the ratios of viscosities and flow rates of the liquids, the width of the channel, and the direction of the electric field. The instability is determined by observing the deflection of the interface. The results are repeatable and show good qualitative agreement with previously reported theoretical works. For example, the order of the applied voltage to destabilize the interface is 95 to 1190 V [2]. The results are presented both in dimensionless numbers to give the reader a means to compare with a theory and with dimensions to show the actual channel dimensions, the flow rates, and the applied voltages. One important result is that the critical voltage or the dimensionless electric number E_b is same when the ratio of the flow rates is unity, independent of the actual flow rates. In fact, the critical voltage remains unchanged within experimental errors, even when the liquids are stationary in the channel. Therefore, the observed instability is different than the instability seen in Couette or Poiseuille flow of two superposed viscous fluids in a channel [33]. Lastly, the effect of the direction of the electric field is studied. For the parallel field, ethylene glycol/silicone oils of different viscosities, castor oil/silicone oils, and olive oil/silicone oils are studied, but none of them were destabilized up to 6,000 volts, which is also in agreement with the literature [18] as the permittivity and conductivity ratios fall in a region where the parallel field cannot destabilize the interface.

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