Microdroplet event recognition and volume detection system based on flexible printed circuit electrode

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Abstract. Changes of microdroplet volume and the occurrence of different droplet events will exert a significant influence on droplet-based microfluidics. Droplet volume is closely related to reagent consumption and hydrodynamic resistance while droplet events reflect actual microchannel conditions. Based on current research, this work was dedicated to fabricate a reliable microdroplet sensing system with event recognition and volume detection function. Signal acquisition was on the basis of capacitance coupled detection and the coplanar fork sensing electrode was fabricated by flexible printed circuit (FPC) which was tens of times cheaper than photolithography. Wave peaks will appear while ink droplets traverse over the fork electrode from above. Shape features of waveform were used to recognize some simple droplet events and the period was employed to calculate droplet volume. Through experimental verification, droplet generation and droplet merging events could be recognized and distinguished by the system. In addition, the detection error between the theoretical volume calculated from formula and the actual volume derived from image analysis was less than 0.5 nL. The sensing system possesses the potential to achieve high throughput detection and surely can be employed to recognize more complicated droplet events in the future.

Keywords: Droplet volume; Droplet event; Capacitive sensor; Flexible printed circuit

1. Introduction

In recent years, microfluidic systems have attracted a large number of researchers from various research fields and have quickly become an important interdisciplinary research topic because of their unique superiorities and wide range of potential applications [1]. Microdroplets act as microreactors in many biochemistry experiments, so that the operation and control of droplets becomes an essential problem [2]. The size of droplets is not only representative of the reagent consumption of microreactors, but also an influence factor of the hydrodynamic resistance [3,4] of the microchannel. When droplets enter or exit the channel, the overall flow field of the microchannel will change, which will lead to unpredictable and complex fluid environment. Thus, it will make a significant difference if we can capture the real-time data of the size or volume of microdroplets. Especially for some integrated microfluidic systems which contain abundant functional units, for example, sorting unit [5], merging unit [6,7], splitting unit [8,9] and so on.

The detection methods can be divided into three classes according to physical characteristics which are optical characteristics, mechanical characteristics and electrical characteristics. Optical characteristics detection is one of the most classical and widely used mean to measure droplet size or volume. Nguyen and others [10] measured droplet size by a laser platform that was integrated into the
microfluidic system. A laser beam traversed the microchannel with running droplets and the location signal will be captured by a photo diode acting as a laser receptor. Chang and others [11] realized droplet sizing by spray imaging and image analysis. Although optical detection is more visualized and high throughput, relatively expensive components and huge body are always restrictions and limitations that prevent it from personalized applications. Furthermore, continuous image capture and analysis requires extremely strong computation system and will make it difficult to secure the real-time ability [12]. Resonance frequency detection is a typical and ingenious mean of mechanical characteristics detection methods. Lee and others [13] detected droplet size by a pulled microcapillary tube resonator of which the resolution can be up to 32 nm. Obviously, droplet frequency should be as low as possible to ensure that there is only one droplet locating in the sensing area at the meantime. Electrical characteristics detection has significant superiority in cost and device complexity over other detection methods with the development of semiconductor and transducer. Previously, lots of researches showed that microdroplet size can be measured by resistance or conductivity change [14]. However general resistance measurement is based on contacted detection which may cause electrolysis or cross contamination. While contactless detection based on capacity coupled technology [15] can avoid these inherent problems fundamentally although there are still some shortcomings during its actual application.

In order to realize droplet sizing as simple as possible and ensure the reliability of system, we adopt electrical detection on the basis of contactless measurement. To reduce external interference, coplanar electrode [16] is a better choice for us compared to other electrode. In this paper, we employed flexible printed circuit (FPC) to fabricate our coplanar electrode which made it tens of times cheaper than photolithography [17]. Real-time capacitance data could be captured while the electrode was bounded beneath the microchannel. Then we analyzed amplitude and period of capacitance signal waveform. Different from traditional detection based on amplitude [18], we creatively employed the period feature to evaluate microdroplet volume which could overcome the interference from background noise or stray capacitance. Besides, shape features of waveform were used to recognize simple droplet events.

2. Materials and methods

2.1. Electrode design

The sensitivity of droplet size measurement is closely relevant to the geometry and parameters of coplanar electrodes. There are two common relative location sensing methods, as figure 1(a) and figure 1(b). Figure 1(a) shows a sensing electrode like an off-state copper. However it may be of significant difficulty to align the electrode interval with the microchannel in the scale of micro meters. And figure 1(b) is a differential structure which can be up to a smaller size. Based on figure 1(b), we designed new structures to acquire as much electrical information as possible, as shown in figure 1(c) and figure 1(d). Figure 1(c) is a popular structure called finger electrode or interdigital electrode [19], which is widely used in projects and experiments. To avoid waveform superposition, we designed a fork electrode as figure 1(d) smaller than finger electrode, and the excitation terminal electrode half surrounded the pick-up terminal electrode to capture more information about droplet inner material.

Generally, coplanar electrodes is always fabricated by photolithography. In our work, we employed flexible printed circuit (FPC) to fabricate the fork electrode which made it tens of times cheaper than the former. In addition to cost saving, FPC fabricating is also very simple without considering the complex microfabrication process. According to previous findings, detection efficiency was mainly influenced by some parameters which were electrode width(W), parallel length of electrode(L), electrode gap(G)[16]. Based on above factors, our fork electrode detection worked as figure 1(e), in which W1 is 125 um, W2 is 175 um and G is 100 um. Because the permittivity of water(78.36 F/m, 25 °C) is dozens of times that of the mineral oil(2.1 F/m, 25 °C), wave peaks will appear while ink droplets traverse over the top of our fork electrode. The substrate and passivation layer of FPC is polyimide so that it can secure the electric insulation. In our work, the thickness of copper is 12
um as thick as the passivation layer. Besides, the whole thickness of the FPC is thin to 100 um so as to guarantee the light transmittance which will be of great convenience for microscope observation.

![Figure 1](image)

**Figure 1.** Different electrode structures fabricated by FPC. (a) Off-state electrode. (b) Differential structure electrode. (c) Finger electrode. (d) Fork electrode. (e) Schematic of capacitance detection on the basis of fork electrode.

2.2. Fabrication of devices

On the whole, the system consists of two parts: droplets generator and droplets sensing system. We employed flow focusing geometry [20] which were widely used in microfluidics to generate uniform droplets. First of all, we fabricated our microfluidic chip(figure 2(a)) carrying droplets generation microchannel by soft lithography. The width of the channel in sensing area is 300 um, three times that of the flow focusing channel. After that, we tried to construct our microfluidic chip with electrodes.

![Figure 2](image)

**Figure 2.** (a) Machining schematic of microfluidic chip with electrode and PDMS focusing geometry. (b) The photo of the final microfluidic chip. (c) Workflow diagram of the complete microdroplet sensing system.

As figure 2(a) display, a piece of FPC was firstly immobilized on a glass slide, and then an as thin as possible poly(dimethylsiloxane-co-(3-aminopropyl)methylsiloxane)(PDMS-linker) film [21] was coated on the surface of FPC to meet the oxygen plasma bounding requirements. For the reason that the material of FPC is polyimide which is difficult to bound with PDMS solidly, the surface modification is necessary. After half an hour of waiting, clean the surface with isopropanol to get rid
of the free molecules. Finally, we bounded it with the flow focusing geometry by oxygen plasma machine to fabricate the final microfluidic chip (figure 2(b)). Based on microfluidic chip, we constructed our droplets sensing system as shown in figure 2(c). The water phase and oil phase are driven by syringe pump that is featured by constant flow to generate droplets. While water-in-oil droplets travel across the sensing area, the sensor circuit will capture capacitive signals by capacitive sensor (AD7150, Analog Devices). AD7150 has pretty short response time (5 ms per channel) as well as very high resolution (1 fF) so as to satisfy some weak signal detection and high-throughput droplets measurement. Besides, the data communication is controlled by a microcontroller (STM32F207, STMicroelectronics). To acquire weak signals, we employed radio frequency cables (RG174, Eastsheep, Beijing, China) to transmit capacitive signal. After acquiring capacitive signals, the capacitive sensor converts analog signals into digital data and send them to the upper computer by serial port or wifi communication. The upper computer can receive and display capacitance data and store them for further analysis by MATLAB.

2.3. Reagent preparation

In this work, we prepared mineral oil as continuous phase and ultrapure water as dispersed phase. In order to guarantee droplet stability, we adjusted the Hydrophile-Lipophile Balance (HLB) with two kinds of surfactants: Span 80 (Aladdin, Shanghai, China) and Tween 80 (Aladdin, Shanghai, China). Then, we mixed 39.9 g mineral oil with 2.5 g above surfactant mixture and followed by 10 minutes vortex shaking. After that, sterile filter unit with membrane filters (Millex, Shanghai, China) was used to purify the continuous phase and dispersed phase. The aperture of the filters for mineral oil is about 0.45 μm while that for water is about 0.22 μm. No matter continuous phase and dispersed phase should be removed air bubbles with ultrasonic cleaners for 30 minutes. Furthermore, poly[dimethylsiloxane-co-(3-aminopropyl)methylsiloxane] (Sigma-Aldrich, America) was employed as PDMS-linker to modify the surface of FPC.

3. Results and discussion

3.1. Event recognition

According to the capacitive signal data acquired by our fork electrode and sensing circuit, we found an interesting phenomenon that different events can be distinguished and recognized by the shape features of waveform. While continuous droplets are generated and travel across the sensing area, periodic signals arise as the curve in figure 3(a) display. For the convenience of observation, we processed the periodic signals by MATLAB as shown in figure 3(a) in which the upper curve and lower curve represent the upper envelope and lower envelope respectively. We can see that the upper envelope is similar to the lower in waveform although their base lines are different. The signal amplitude increases while a droplet moves close to the sensing area and decreases with its leave. Therefore, a common droplets generation event can be confirmed while the periodic signals like the curve in figure 3(a) appear. The waveform features can be summarized as follows: obvious amplitude variation, periodicity, symmetry and so on.

Although there are many similar characters between figure 3(a) and figure 3(b), the curve in figure 3(b) show a special feature which can be used to distinguish events different from figure 3(a). The box in figure 3(b) shows a sharp rise on the left edge of the periodic signals while a slow descent on the right edge. When a droplet traverses the normal sensing area like figure 3(c), obvious amplitude arise will appear but not a cracked peak because of the short enough response time of AD7150 unless an emergency event happens. Through waveform analysis and microscope observation, we found that the emergency event happening in figure 3(b) is a droplets merging event other than a common droplets generation event in figure 3(a). A rough channel environment like figure 3(d) is easy to cause the droplet merging events. Therefore, a periodic droplets merging event can be confirmed while the periodic signals like figure 3(b) appear. In addition to periodicity and obvious amplitude variation, the most typical feature is the asymmetry accompanied by a cracked change of the waveform.
Figure 3. (a) Capacitance signals and envelope waveforms of droplet generation events. (b) Capacitance signals of droplet merging events. (c) Droplet generation events of normal channel with different electrode structures: 1 fork electrode; 2 finger electrode; 3 off-state electrode. (d) Droplet merging events of rough channel(1-3).

In order to evaluate the above recognition method, we have statistics on the experimental results. Figure 4 is the relationship line between recognition rate and droplet periods. We can see that the recognition rate is positively related to the periods and it approaches 1 as the period increases.

Figure 4. The relationship between recognition rate and droplet periods.

3.2. Droplet volume detection

Generally, electrical detection of droplet volume is based on the magnitude of the signal amplitude. Research has shown that there is a linear relationship between them theoretically [18]. However, weak capacitive signal is extremely easy to mix in noise which can lead to a wrong value of the amplitude. Besides, the linear relationship between the droplet size and the signal amplitude will distort while more than one droplet exist in the sensing area, so that it can only satisfy some low-throughput and low flow rate applications. In our work, we tried to employ the period other than signal amplitude to
evaluate microdroplet volume which could overcome the interference from background noise and the limitations about throughput and flow rate. Because the syringe pump is featured by constant flow, droplets generation should satisfy the following formula:

\[ V = QT \]  

(1)

where Q is the flow of dispersed phase, \( V \) is the volume of the droplet and \( T \) is the detected period of a droplet. According to the above formula, we can find the theoretical linear relationship between droplet volume and period under the constant flow(Q). The period of droplets can be calculate from capacitive signal in figure 3(a) and figure 3(b). Besides, the noise mixed in the signal is difficult to have a substantive influence on the period because the great amplitude variation makes it easy to distinguish every period. We just need to calculate the time span and evaluate the droplet volume according to formula(1).

![Figure 5. The relationship between droplet volume and detected droplet periods.](image)

In order to verify formula(1), we calculated droplets volume with different periods as shown in figure 5 in which the squares referred to actual volume derived from image analysis and the triangles referred to theoretical volume derived from formula(1). Because droplets with right size can be approximated as spheres or columns, we calculated the approximate values of droplet volume by ImageJ to set comparison for theoretical volume. To acquire different periods, the flow of discrete phase remained at 30 \( \mu L/h \) while the continuous phase increased from 20 \( \mu L/h \) to 70 \( \mu L/h \) at the rate of 10 \( \mu L/h \). The line in figure 5 is the fitting straight-line of approximate actual volume. We see that the calculated values agree with the theoretical values roughly from the fitting straight-line and formula(1). Therefore we can calculate the theoretical volume of a droplet by formula(1) to represent the actual volume on the condition that the flow of dispersed phase was known. Compared to image analysis, this method can overcome the influence of droplet deformation and irregular channels. According to figure 5, the detection error was less than 0.5 \( nL \) and the time resolution was as low as 5 ms thanks to the high performance sensor.

4. Conclusion

To sum up, we fabricated a reliable microdroplet sensing system based on capacitance coupled detection and verified its performance in droplets volume detection and events recognition. The coplanar fork sensing electrode of microfluidic chip was fabricated by FPC which made it a lot of cheaper. Besides, we chose a commercial sensor with shorter response time and higher resolution to capture capacitance signal. The sensing system can distinguish different events according to the shape features of waveform accurately as well as measure droplet volume on the basis of measured period. In addition, the theoretical formula was verified by a syringe pump with known dispersed phase flow. Because of some limitations, the events recognition was just verified by simple events: droplets generation events and droplets merging events. In the future, the microdroplet sensing system surely
can be used to recognize more abundant and more complicated droplet events. Besides, detection based on period possesses the potential to achieve high throughput detection compared to other electrical detecting methods.

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