Precise detection of the size of monodisperse droplets considering the fluctuations induced by the droplet generation process

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Abstract
For droplet microfluidics, the electrical-detection method which can precisely detect the size of monodisperse droplets is demonstrated in this paper. In a Flow-focusing microdroplet generator, three pairs of the microelectrodes are allocated along the microchannel, and during the passing-by process of each droplet, both the length, the velocity and the production speed of the droplets can be obtained from the experimental measurements of the time-varying capacitance between each pair of the microelectrodes. Particularly, for different geometries of the Flow-focusing microchannel, the method of the electrical-detection is validated experimentally over a wide range of the typical conditions of monodisperse droplet production. In addition, the droplet size measured by the electrical-detection method is compared with that by the method of image processing, and the detection precision of the electrical-detection method is verified experimentally. Most importantly, by calculating the root-mean-square value of the droplet lengths for three pairs of the microelectrodes, the detection precision of the droplet size can be increased drastically.

Keywords: droplet microfluidics, electrical detection, size, precision

1. Introduction
Droplet microfluidics are widely applied for interdisciplinary research such as particle synthesis[1-4], biological analysis[5-9] and cell detection[10,11]. In particular, how to precisely and efficiently test the size of the droplets is quite important for droplet microfluidic systems[12-14]. To date, as monodisperse droplets are flowing along the microchannel, the droplet size is mostly measured by the method of image processing[15-17]. However, to the best of our knowledge, the process of the image processing is quite complicated and time consuming[18], which can affect the dynamic characteristics of the droplet microfluidic systems[19-21]. Especially, in order to ensure the detection accuracy of the droplet size, the images of droplets with high resolution need to be captured by some expensive and sensitive detection instruments including the microscope and high-speed camera, which can increase the cost of the detection devices.

To simplify the detection process of the droplets and lower the expense of the detection devices, the method of electrical detection has been applied to measure the droplet size for droplet microfluidic systems[22,23]. Particularly, the capacitance between each pair of the microelectrodes can be calculated based on the principle of the electrical-detection method, and by measuring the time-varying capacitance while individual droplet is passing through the microelectrodes, a linear relation between the
droplet length and the magnitude of the capacitance variation can be obtained approximately\(^{[22]}\). In particular, compared with the method of image processing\(^{[18]}\), the detection process is greatly simplified and the detection speed can be increased significantly for the electrical-detection method. However, the detection precision of the electrical-detection method is only discussed qualitatively, and the measuring accuracy of the droplet size needs to be studied quantitatively.

In this paper, the droplet microfluidic electrical-detection device is established and the electrical-detection method is verified experimentally for a wide range of the droplet velocity, the droplet size and the microchannel geometries. Different pairs of the microelectrodes are allocated along the Flow-focusing microchannel, and by measuring the time-varying capacitance between each pair of the microelectrodes during the passing-by process of each droplet, both the length and the velocity of the droplets can be calculated. By comparing the droplet size measured by the electrical-detection method with that by the method of image processing, the detection accuracy of the droplet size can be quantified. Additionally, the effects of the droplet production process on the detection precision of the droplet size are also discussed. Most importantly, by calculating the root-mean-square value of the droplet lengths measured by three pairs of the microelectrodes, high detection precision of the droplet size can be achieved for the droplet microfluidic electrical-detection device.

2. Experimental setup

The droplet microfluidic electrical-detection device is established for measuring the size and velocity of the droplets. The working principle of the droplet microfluidic electrical-detection device is shown in Fig. 1.

Fig. 1 Working principle of the droplet microfluidic electrical-detection device

As monodisperse droplets are flowing along the microchannel and passing by the detection microelectrodes, the detection process of the size of each droplet is shown in Fig. 2.
Here, the microelectrodes are fabricated and integrated with the Flow-focusing microdroplet generator. As each droplet is passing through the microelectrodes, the location between the droplet and the microelectrodes is changing, and meanwhile, the capacitance between each pair of microelectrodes is a function of time. Especially, during the passing-by process of each droplet, the maximum magnitude of the capacitance variation can be obtained while the gap between each pair of the microelectrodes is fully filled by the droplet.

For the experiments of droplet generation, the silicone oil is chosen as the continuous phase and the DI water is chosen as the dispersed phase. The viscosity of the DI water and silicone oil is $\mu_d = 1 \text{cP}$ and $\mu_c = 20 \text{cP}$, respectively. The dielectric constant of the DI water and silicone oil is $\varepsilon_d = 80$ and $\varepsilon_c = 2.5$, respectively. By controlling the flow rates of the two phases, both the size and the velocity of the droplet can be verified. Particularly, a Flow-focusing microchannel is designed to generate monodisperse droplets, and different widths of the microchannels are selected to study the detection precision of the electrical-detection method. For our experiments of droplet production, the widths of the microchannels can range from 50 to 300 $\mu$m.

3. Mathematical model

In this paper, individual droplets are formed in the Flow-focusing microchannel and the sizes of droplets are measured by the method of electrical detection. As each droplet is passing through the microelectrodes, the distribution of the electrical field between the microelectrodes can be analyzed simultaneously, as shown in Fig. 3.
According to the literature\(^{[22]}\), for each pair of the microelectrodes designed here, the capacitance between a pair of microelectrodes can be described by

$$C = \frac{2\varepsilon_0\varepsilon_r w_e}{\pi} \ln \left[ 1 + \frac{2w_e}{d} + \sqrt{\left(1 + \frac{2w_e}{d}\right)^2} - 1 \right]$$

(1)

where \(w_e\) is the width of the microchannel, \(\varepsilon_0\) is the electric constant, \(\varepsilon_r\) is the dielectric constant and \(w_e\) is the effective width of the microelectrode.

Here, the water droplets are formed in the Flow-focusing microchannel. Since the dielectric constant of water is nearly 30 times of that of silicone oil, while the droplet is passing through the microelectrodes, the capacitance between each pair of microelectrodes can be increased significantly. Based on the equation (1), the magnitude of the capacitance variation induced by each droplet can be calculated

$$\Delta C = k_c w_e$$

(2)

We note that while each droplet is passing by the microelectrode, the amplitude of the capacitance variation is proportional to the width of the microchannel. Especially, the proportional gain coefficient can be determined by

$$k_c = \frac{2\varepsilon_0 (\varepsilon_d - \varepsilon_e)}{\pi} \ln \left[ 1 + \frac{2w_e}{d} + \sqrt{\left(1 + \frac{2w_e}{d}\right)^2} - 1 \right]$$

(3)

where \(\varepsilon_d\) is the dielectric constant of water and \(\varepsilon_e\) is the dielectric constant of silicone oil.

Actually, the theoretical value of capacitance between the microelectrodes is a function of time during the passing-by process of each droplet. In particular, while the droplet is fully across each pair of the microelectrodes, the maximum value of the capacitance can be reached for each pair of the microelectrodes. Here, we define \(\Delta t\) as the time interval of the capacitance pulse, and based on the geometrical parameters of the microelectrodes, the droplet length can be calculated

$$L_d = w_e + d + V_d \Delta t$$

(4)

where \(V_d\) is the velocity of the droplet. Additionally, based on the distance \(S_d\)
between two pairs of the microelectrodes and the time interval $\Delta t_d$ of the droplet passing through the two pairs, the velocity of the droplet is given by $V_d = S_d / \Delta t_d$.

4. Results and discussion

To validate the mathematical model of the droplet microfluidic electrical-detection device, different widths of the microchannels are designed for the experiments of droplet generation, and various lengths of droplets can be produced in the Flow-focusing by varying the flow rates of the two immiscible fluids. Based on the mathematical model, as different lengths of droplets are passing through the microelectrodes, the capacitance between each pair of the microelectrodes is a function of time. For Flow-focusing 1, the width of the microchannel is specified as $w_c = 100 \, \mu m$, and by producing different sizes of the droplets in microchannel, the time-varying capacitance between the microelectrodes is measured experimentally, as shown in Fig. 4.

![Fig. 4](image.png)

a) $L_d = 210 \, \mu m$  
b) $L_d = 330 \, \mu m$

Fig. 4 The capacitance of the microelectrodes as a function of time while different lengths of droplets are passing through the microelectrode for Flow-focusing 1

For Flow-focusing 2, the width of the microchannel is specified as $w_c = 150 \, \mu m$, and while monodisperse droplets are formed in the microchannel, the time-varying capacitance between the microelectrodes is measured experimentally, as shown in Fig. 5.
Fig. 5 The capacitance of the microelectrodes as a function of time while different lengths of droplets are passing through the microelectrode for Flow-focusing 2

For Flow-focusing 3, the width of the microchannel is specified as $w_c = 200 \mu m$, and while monodisperse droplets are generated in the microchannel, the time-varying capacitance between the microelectrodes is measured experimentally, as shown in Fig. 6.

Fig. 6 The capacitance of the microelectrodes as a function of time while different lengths of droplets are passing through the microelectrode for Flow-focusing 3

From the experimental results, it can be observed that the capacitance of the microelectrode is a periodic function of time. Especially, a pulse signal of the capacitance can be generated during the passing-by process of each droplet. More importantly, the pulse amplitude of the capacitance variation is mainly determined by the geometrical parameters of the microchannel and independent of the droplet size. Additionally, the pulse width of the capacitance variation is rising obviously as the length of the droplets increases.

In order to test the linear relation between the magnitude of the capacitance variation and the width of the microchannel, the time-varying capacitance during the passing-by process of each droplet is measured for different widths of the microchannel. Especially, as the microchannel width ranges from 50 to 300 $\mu m$, the magnitude of the capacitance
variation can be quantified based on the experimental measurements, which are compared with the theoretical predictions obtained from the mathematical model, as shown in Fig. 7.

![Fig. 7](image)

**Fig. 7** Comparison between the experimental measurements and the theoretical predictions of the magnitude of the capacitance variation during the passing-by process of each droplet. The error bar represents the standard deviation of five separate measurements for a given width of the microchannel.

From Fig. 7, we note that the measured magnitude of the capacitance variation is approximately a linear function of the width of the microchannel. Especially, good agreements are found between the experimental measurements and the theoretical predictions as the microchannel width spans from 50 to 300 µm. As a result, the mathematical model of the droplet microfluidic electrical-detection device can be verified experimentally.

Based on the mathematical model of the electrical-detection device, the velocity of the droplets needs to be measured so that the length of the droplets can be calculated from the experimental results. To ensure the measuring precision of the droplet speed, both the methods of electrical detection and image processing are used to test the velocity of the droplets. Here, \( V_d' \) represents the droplet speed obtained by the method of electrical detection and \( V_d \) represents the droplet speed acquired by the method of image processing. Consequently, the experimental measurements of the droplet velocity tested by the electrical-detection method are compared with that by the image processing method, as shown in Fig. 8.
Fig. 8 Comparison between the measured velocity of the droplets by both the methods of electrical detection and image processing. The error bar represents the standard deviation of five separate measurements for a specified flow condition.

From the experimental results, it can be observed that the velocity of the droplets obtained by the two methods are consistent with each other. Therefore, the droplet length can be calculated from the experimental measurements of the droplet speed. However, based on our formal study\cite{23}, there are periodic fluctuations which are induced by the dynamic process of droplet generation, and consequently, the velocity of the droplet is varying with time during each period of droplet formation. Here, $\langle V_d \rangle$ is defined as the average speed of individual droplet for each droplet formation period, and $V_d(t)$ is defined as the time-varying droplet velocity. As $V_d(t)$ is divided by $\langle V_d \rangle$, we can obtain the experimental measurements of the periodic velocity fluctuations induced by the droplet production progress, as shown in Fig. 9. We note that the droplet velocity $V_d(t)/\langle V_d \rangle$ is changing periodically with time, and the testing accuracy of the droplet size can be affected by such periodic fluctuations.

Fig. 9 Experimental measurements of the periodic velocity fluctuations induced by the process of droplet formation. Here, the time-varying droplet velocity $V_d(t)$ is divided by the average speed
\langle V_d \rangle$ of the droplet for each droplet formation period.

In order to reduce the influences of the periodic fluctuations on the detection accuracy of the droplet size, three pairs of the microelectrodes are allocated along the microchannel (see Fig. 2), and while each droplet is flowing along the microchannel, the droplet length is tested by the three different pairs of the microelectrodes independently. In particular, to verify the detection precision of the droplet size, the length of the droplets tested by the electrical-detection method are compared with that by the image processing method, as shown in Fig. 10. Here, $L_d'$ represents the droplet length obtained by the method of electrical detection and $L_d$ represents the droplet length acquired by the method of image processing.

![Fig. 10 Comparison between the measured length of the droplets by both the methods of electrical detection and image processing.](image)

From the above results, we note that the droplet length measured by the electrical-detection method coincides with that of the image processing as the length of the droplets spans from 100 to 500µm. In particular, as the flow rates of the two immiscible fluids are specified for the Flow-focusing microdroplet generator, the theoretical value of the droplet length can be assumed as a constant. However, by calculating the ratios of $L_d'/L_d$, a small deviation can be observed between the droplet length $L_d'$ and $L_d$. During droplet generation, because of the periodic pressure fluctuations which are induced by the dynamic process of droplet formation, the detection precision of the droplet size can be affected by the instability of the droplet production progress.

In addition, the root-mean-square value $L_{d\text{rms}}$ of the droplet length can be calculated from the experimental measurements of three different pairs of the microelectrodes, which is compared with $L_d$ of the image processing method, as shown in Fig. 11. By
comparing the experimental results of Fig. 10 and Fig. 11, it can be seen that the deviation between the droplet length \( L_d^{\text{rms}} \) and \( L_d \) is drastically reduced for the electrical-detection method. As a result, while there are periodic fluctuations induced by the droplet production process, the detection precision of the droplet size can be obviously increased by calculating the root-mean-square value \( L_d^{\text{rms}} \) of the droplets.

![Fig. 11 Comparison between the root-mean-square value of the droplet length by electrical-detection method and the droplet length by image processing method. The error bar represents the standard deviation of five separate measurements for a specified droplet size.](image)

From the above discussion, the mathematical model of the droplet microfluidic electrical-detection device can be verified experimentally over a wide range of the droplet velocity, the droplet size and the microchannel geometries. Most importantly, by calculating the root-mean-square value \( L_d^{\text{rms}} \) of the droplet lengths tested by three pairs of the microelectrodes, the detection precision of the droplet size can be improved significantly for the electrical-detection method.

5. Conclusions

The electrical-detection method for droplet-size detection is demonstrated in this paper. In particular, the mathematical model of the droplet microfluidic electrical-detection device is established, which can be verified experimentally over a wide range of the droplet velocity, the droplet size and the microchannel geometries. Based on the mathematical model, both the length and the velocity of the droplets can be quantified by measuring the time-varying capacitance between each pair of the microelectrodes during the passing-by of individual droplet. Compared with the method of image processing which requires expensive detection devices such as microscope and high-speed camera, the testing expense of the droplets can be greatly reduced by the electrical-detection method. Most importantly, by properly allocating three pairs of the microelectrodes along the microchannel, the influences of the droplet generation process on the measuring accuracy of the droplet size are reduced significantly. As a
result, high detection precision of the droplet size can be achieved for the droplet microfluidic electrical-detection device.

**Declarations**

**Availability of data and materials**
The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Competing interests**
The authors declare no conflict of interest.

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**Authors' contributions**
Wen Zeng: Writing-original draft, Writing-review and editing, Methodology, Conceptualization, Validation, Supervision.
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