Coalescence and Separation of Monodisperse Droplets

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Abstract. The liquid ejected from a small hole will break out under the action of the surface tension. If an excitation is applied with a certain frequency, the jet will disintegrate into droplets, and the droplets flowing with uniform particle size and space will be obtained, which can be used for study of droplet dynamics. In the process of developing a droplet generator, it is found that the different methods of liquid supply have significant effects on the droplet flow quality. The droplet flow will catch up to coalesce or separate. By using the dimensionless parameters Reynolds number, Weber number, Bond number and Ohnesorge number to analyze the process of the droplet flow, the drag coefficient $C_d$ of the droplet in the process of catching up is obtained. The role of inertial force, surface tension and viscosity effect in the process of droplet coalescence and separation is described, and the device is improved to prohibit this phenomenon happening.

Introduction

Droplet generation devices capable of producing uniform particle sizes are widely used in industrial applications such as granulation production[1, 2], rapid prototyping [3] and even space applications[4, 5], coating on substrates, as well as particle manufacturing [2], requiring the use of controllable particle size droplet forming techniques, and they are also used to study the energy transport processes, such as the coalescence of two droplets, droplets against the solid surface or liquid film surface or droplet evaporation.

In 1829, Bidone studied the form of a jet when the liquid passed through different shapes of orifices. The basic principle of the monodisperse droplet generating device is the controlled Rayleigh-type jet break-up, which stimulates the liquid jet by applying excitation to the liquid ejected from a round orifice or nozzle. In 1950, Dimmock developed an early uniform droplet generator working with vibrating capillaries, which was published on Nature.

The researchers hope that through more stringent parameter design to obtain the desired monodisperse droplets. Ashgriz etc, studied the impact of external vibration waveform on the liquid column fracture. Mansour used nonlinear analysis to compute the collapse of elongated satellites which showed short waves propagating on their surfaces.

Researchers in the field of aviation hope to study the effects of Supercooled Large Droplet (SLD) on wing icing, particle sizes ranging from 100um to 500um, especially droplets of 200um-300um particle size, it’s rare in the previous work.

The uniform droplet generation device belongs to the category of atomization and spray fields, and the researchers have focused on the uniformity of the droplet size and spacing, while there is not much study of the nonuniformity. When the droplet spacing is not equal, the droplets will catch up to coalesce or separate. The study shows that this is the result of interaction between inertial force, surface tension and viscosity effect.

To prohibit the phenomenon of droplet catch-up coalescence is necessary for designing a monodisperse droplet generator. Both the liquid supply mode and excitation signal were improved to make sure that monodisperse droplets with uniform particle size and space can be obtained in this work.
Droplet Generating Device

Working Principle

Controlled Rayleigh-type jet break-up principle, that is, the dynamic instability of the droplet (jet) under the action of the surface tension, as shown in Figure 1, when the excitation signal of a certain wavelength is applied to the jet, the jet will decompose into droplets of uniform size. The droplet size depends primarily on the size of the orifice and can be adjusted by pressure, excitation signal, and so on. Piezoelectric transducers (piezoelectric ceramic sheets) are used to generate interference waves on the jet and to control the generation of satellite droplets (small droplets).

\[ \omega_j^2 = \sigma_0 \frac{ka}{\rho} (1 - \kappa^2 a^2) \frac{I_0(ka)}{I_1(ka)} \tag{1} \]

In this relation, \( \omega_j \) is a growth rate of a disturbance, \( \sigma_0 \) and \( \rho \) are the surface tension of the liquid against the ambient gas and density of the liquid, respectively, and \( I_0 \) and \( I_1 \) modified the first kind of Bessel functions, make \( \alpha = \omega_j / \omega \) the perturbation wave growth rate function image is shown in Figure 2, which shows that for the liquid jets studied in the present work their growth rates can be approximated by the linear growth rates of free inviscid jets.

Schneider and Hendricks found the suitable range of wave numbers through experiments as \( 0.45 \leq \kappa \leq 0.95 \), at that time, the jet is able to produce a stable monodisperse droplet. Brenn considered a more precise range \( 0.3 \leq \kappa \leq 0.9 \).

The wavelength is expressed by the dimensionless parameter \( We \) and the frequency of the excitation signal, and the frequency range of the excitation signal can be obtained as follows:

\[ 0.3 \leq \kappa = \frac{\pi}{\sqrt{We}} \sqrt{\frac{\rho d^{3/2}}{\sigma}} f \leq 0.9 \tag{2} \]

Walzel and Ohnesorge experimentally concluded that controlled Rayleigh-type jet break-up stability in practical situations is defined by the inequality:

\[ 14.5 Oh^{0.08} \leq We \leq 2345.46 Oh^{0.412} \tag{3} \]

In this condition, \( Oh = \mu_d / \sqrt{\rho \sigma d} \), \( We = \rho d^2 / \sigma \), \( \mu_d \) is the droplet viscosity coefficient, \( \rho \) is the droplet density, \( d \) is the orifice diameter, \( U \) is the jet velocity.

With the excitation frequency \( f \), the diameter range of the droplets is got:

\[ D = \left( \frac{3 \pi}{2} \kappa^3 \right) d = \left( \frac{3}{2} \sqrt{\frac{\sigma}{\rho d^{3/2}}} \right) d \tag{4} \]
$D$ is the droplet's diameter, the liquid through the small holes to generate a jet need to overcome a certain resistance, the minimum working pressure of the device is \( P_{\text{min}} = \frac{32\mu hU}{D^2} \), \( h \) is the thickness of the orifice, the actual working pressure will be greater than this pressure. From the formula, the pressure is inversely proportional to the square of the diameter, if the aperture is very small, the required pressure will be very big.

**Experimental Devices and Parameters**

The droplet generating device is shown in Figure 3

![Figure 3. The system of droplet generator.](image)

The launch tube is one of the most essential parts of the droplet generating device, as shown in Figure 4, which is a sectional view of the droplet launching tube.

The experimental parameters are as follows:

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hole diameter</td>
<td>0.25</td>
<td>mm</td>
</tr>
<tr>
<td>Average flow</td>
<td>8.837</td>
<td>mL/min</td>
</tr>
<tr>
<td>Pressure</td>
<td>0.1</td>
<td>Mpa</td>
</tr>
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<td>Waveform</td>
<td>Sinusoidal wave</td>
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<tr>
<td>Frequency</td>
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<td>KHz</td>
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<tr>
<td>Voltage amplitude</td>
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<td>V</td>
</tr>
<tr>
<td>Frames</td>
<td>16000</td>
<td>fps</td>
</tr>
</tbody>
</table>

In the course of the experiment, Ultrasonic Cleaning Instrument plus Ethanol is adopted to clean, cleaning 10-15 minutes can remove impurities, while the main body of the launch tube of other parts are also cleaned to prevent secondary contamination. Figure 5 shows the exit of the launch tube, jet enjects from the orifice with the effect of pressure inside and gravity outside.

![Figure 5. Image of droplet formation at the exit of the launch tube.](image)

Figure 6. Jet image with scaleplate.

In order to obtain the dimensionless parameters, the velocity of the droplet should be gotten first. As shown in Figure 6, the distance between the two frames can be obtained by adding the scale in the
field of view of the high-speed camera. The interval can be used to obtain droplet speed. In order to reduce the reading error, an average speed of 30 frames is adopted as the droplet speed.

Excitation Signal
Using the function signal generator can produce different frequency of rectangular wave, sinusoidal wave or triangular wave, and even can be obtained above the superposition of the waveform, but the function of the signal generator output voltage is not enough to directly drive the piezoelectric ceramic vibration, therefore, a power amplifier device is needed. In the experiment, the sinusoidal wave with different frequency is used as the excitation signal, and then rectangular wave signal is adopted. As a result, rectangular wave signal is more likely to produce the uniform droplet flow than the sinusoidal wave signal.

Liquid Supply Mode
In order to break-up through controlled Rayleigh jet to produce monodisperse droplets, the liquid must be supplied to the launch tube at a constant flow rate. The experiment takes a total of three liquid supply modes:

1. Using a peristaltic pump for the liquid supply, the advantage of this way is that the pulsation is smaller than gear pump, the liquid flow range is larger than the injection pump, the liquid supply is stable. The disadvantage is that the pressure is small, when the working pressure increases, an occurrence of backflow phenomenon may happen, when the aperture is between 0.15mm-0.25mm, such liquid supply is adopted;

2. Using an air compressor plus liquid storage tank for the way, the advantage is almost no pulsation, the liquid is stable, the liquid pressure is larger than the peristaltic pump, working range of 0Mpa-0.8Mpa, the flow rate is not limited by the range. This way is ideal, but because the impurities in the tank or dust in the air may easily obstruct the orifice. When the orifice size is between 0.075mm-0.25mm, such liquid supply is used;

3. Using injection pump for the liquid supply, the advantages are pure, with no impurities, pulsation is smaller than peristaltic pump, the liquid is stable, it can provide a small flow of liquid, but the liquid can’t be continuous, besides, syringe pressure is not enough high, when the working pressure reaches 0.4Mpa, plastic syringes will be a serious deformation, the glass syringe will overflow. When the pore size is in the range of 0.075 mm- 0.15 mm, such liquid supply is adopted.

Droplets Catch Up

Analysis of Droplet Force
There are many factors that may affect droplet behavior, such as the following:

1. Pneumatic resistance;
2. droplet size;
3. droplet velocity, associated with air;
4. droplet acceleration, or gravity;
5. air density and viscosity;
6. droplet density and viscosity;
7. surface tension;

Since the shape of the droplet changes reasonably slowly during the movement, and there is no significant internal flow, then the droplet viscosity could be ignored, according to Buckingham Pi theory. Finally, the dimensionless parameter Re, Wb, Bo, Oh are used as the possible dimensionless numbers.

When the droplet is spherical or fixed shape, the surface tension does not work, usually using the Reynolds number and Drag coefficient Cd to describe the aerodynamic drag force. Drag coefficient Cd can be obtained using empirical formula. Therefore, if study the force and movement state of droplets in the process of catching up, mainly use Re and the Drag coefficient Cd to describe.

Then, when the droplets collide, an occurrence of coalescence or separation, the shape of the droplets changed significantly, need to consider the impact of surface tension, as a result, the number of Bo is taken into consideration.

The dimensionless parameters and the Drag coefficient are as follows:
\[ \begin{aligned} Re &= \frac{\rho U D}{\mu} \\ We &= \frac{\rho U^2 D}{\sigma} \\ Bo &= \frac{\rho g D^2}{\sigma} \\ Oh &= \frac{\mu_s}{\sqrt{\rho_s \sigma D}} \\ Cd &= \frac{8F}{\pi D^2 \rho U^2} \end{aligned} \] (5)

\[ Cd = \begin{cases} 24 / Re & Re \leq 1 \\ 24 \times (1 + Re^{0.67}) / Re & 1 < Re \leq 1000 \\ 0.44 & Re > 1000 \end{cases} \] (6)

The parameters in the formula are as follows:

For the above formula, add two notes:

(1) the droplet characteristic diameter \( D \) is the equivalent diameter \( D \) when the droplet is spherical, and when the droplet deformation ratio is high, the diameter of the widest part of the droplet or the thickness \( h \) of the droplet can be used as the characteristic diameter;

(2) When considering the deformation and resistance of droplets, the parameters of ambient air are calculated as characteristic parameters, the surface tension coefficient is constant.

According to experimental observations, the velocity of the droplet is between 2.84 m/s to 2.93 m/s, and the dimensionless flow parameters are as follows: the Reynolds number is 90, the number of Weber is 54, the number of Ohnesorge is 7.43E-3, Bond number is 0.03, Drag coefficient \( Cd \) is 1.147.

### Analysis of Droplet Motion

**The Droplets Catch Up and Collide.** When the droplet spacing is uniform, droplet Drag coefficient is the same, as the droplet spacing becomes smaller, the Drag coefficient decreases. Because of the ambient airflow or disturbance, droplet in the stream moves to the downstream direction a little bit, it will increase the resistance of the upstream droplets and reduce the resistance of the downstream droplets, the result is to further increase the displacement of the droplets, resulting in uneven spacing of droplets, often as pairs, or triplets.

The effect of the air flow between the droplets, the droplet spacing and the Reynolds number, have an effect on the drag coefficient. Generally speaking, when the distance between the two droplets is appropriate, vortex between them makes droplets move closer to each other. For example, when the spacing is twice of the droplet diameter, there could be negative drag on the downwind drops. In a stream of spherical droplets the interactions could be complex where they are unevenly spread and misaligned and no data was found for that situation.

Figure 7 shows the calculation of the external flow field of the droplet. In the linear shear flow, the difference between the spherical droplet and the rigid sphere is small and can be neglected, therefore, assuming that the droplet surface is a wall, and other boundaries are velocity inlet, outflow, symmetry axis and non-slip wall. The calculated results are shown in Figure 8, where the flow line distribution at \( Re = 90 \) shows that there is a ring vortex between the droplets, this is the main reason for droplet coalescence. Usually, only the spacing reaches 10 to 15 diameters apart, this effect can be neglected.

![Figure 7. Schematic diagram of computational domains.](image1)

![Figure 8. Streamlines at Re = 90 of spherical droplet.](image2)

As shown in Figure 9 (the left side is the direction of gravity), the droplet flow begins to catch up, and the spacing between the left sixth and the fifth droplets begins to decrease.
Experimental Results and Analysis

Double Droplet Catching Coalescence

When droplets in the stream are close to each other and the spacing is twice the diameter, droplets may interact with each other, and droplets may converge, as shown in Figure 10.

Droplets in the droplet chain will oscillate under inertia, which is caused by the deformations upon pinch-off from the jet. After the droplets are fused, this oscillate is more pronounced, like a spring oscillation. The droplet surface tension is analogous to that of the spring vibrator, the aerodynamic is equivalent to external forces, and the viscosity effect represents the damping force. Sometimes, the droplet fusion process occurs very quickly, there is no oscillate on the complete integration, as shown in Figure 10. Sometimes, the fusion process needs to undergo two or three oscillate process to fully coalesce, as shown in Figure 11, in the gravity and vertical gravity direction, respectively, an occurrence of two oscillates, completes the fusion process. The newly generated droplet volume is twice that of the original, aerodynamic force increases, it is more likely to deformation and break-up, which is one of the reasons why it is necessary to prohibit droplet fusion.

Double Droplets Chase and Separate

When the droplets are close to each other, and the fusion occurs, droplet oscillation is sufficient to overcome the inertia force, then the fusion can’t be completed.

As shown in Figure 12, when the droplet (No.2 droplet) is gradually approaching the downstream adjacent droplet (No.1 droplet), the spacing is close to twice the diameter, No.2 droplet is deformed due to the effect of the vortex of No.1 droplet, its upper part is close to No.1 droplet. Under the action of inertia deformation, however, No. 1 droplet separated from the No. 2 droplet, this is the first kind of separation process.

As shown in Figure 13, the following two droplets undergo a contact--separation--re-contact--re-separation process. This time, the droplet surface tension plays a leading role. When the two droplets are in contact with each other due to wake effect, the drops have to keep the smallest surface area under the surface tension, that is, the spherical shape, so that the contact part is narrowed, and the shape of the droplet is changed from spherical to ellipsoid after separation. This time, Drag coefficient becomes larger, the eddy current effect of the droplet increases, re-contact. In the role of
surface tension, however, droplet fusion failed to complete again. After the separation, droplets are restored spherical, which is the second kind of separation process.

**Conclusion**

Droplets occur to catch up and coalesce, obviously will lead to droplet flow spacing uniform, affecting the size of droplets produced, this situation should be prohibited, solutions in the following may help in this situation:

1. Change the liquid supply form, add a constant pressure valve to reduce the pulsation effect of the peristaltic pump;
2. Increase space through reducing the excitation frequency, it do work sometimes but this method has limited ability to increase the spacing, and if the frequency reduces too low, it can’t produce droplets with proper roundness;
3. The third way is to use electrostatic charging, the repulsive forces between the droplets keep them apart and prevent them coalescing. With the help of the pulse generator and power supply, excess droplets can be easily removed, so that the space of droplets left increases, it was used in continuous ink-jet printers;
4. The fourth way is to install a spinning disc at the exit of the tube, thereby only the required droplets are allowed to go through.

In order to prohibit the above-mentioned phenomenon, firstly an air compressor and storage tank should be used to replace peristaltic pump, and a constant pressure valve to control the pressure in the pipeline to maintain a constant value; Then signal waveform is changed, using rectangular wave signal instead of sinusoidal wave signal, and finally uniform particle size and spacing droplets will be obtained, as shown in Figure 14, you can see the smaller the particle size, the closer the droplet shape is to spherical shape, because smaller the particle size, the greater the difference in pressure inside and outside the droplet, and the less affected by the aerodynamic force.

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**References**


