Abstract

We report here, the time-lapse of the breakup phenomenon of a charged droplet (diameter $\sim$100–300 $\mu$m) levitated in an electrodynamic (ED) balance. During the breakup process, a levitated charged droplet undergoes evaporation leading to a reduction in droplet size and increase in the corresponding surface charge density. As the surface charge density reaches to a critical value, known as Rayleigh limit, the droplet undergoes breakup emitting highly charged progeny droplets. All the successive events of the droplet breakup process such as drop deformation, breakup, and relaxation of the drop after ejection of progenies have been recorded using the high-speed camera at 1.3 hundred thousand frames per second. The droplet is observed to eject a 3-5 number of progeny/daughter droplets by end pinching mode of droplet breakup via jet formation. A jet is observed to relax back after ejecting 31% of the total charge and $\sim$2.5% of the total mass. A suitable theory supplements experimental observations and a reasonable agreement is observed between theoretical calculations and the experiments. Additionally, the theory is extended for the calculation of the total mass loss during the lifetime of a charged droplet.

Keywords: Charged droplet, Levitation, Electrodynamic balance

1. Introduction

One often encounters charged droplets in various atmospheric and industrial processes, for example, electrified cloud droplets, electrospray in the context of ion mass spectroscopy, and inkjet printing, etc. Rayleigh [1] first derived the threshold charge at which the repulsive electrostatic force equals or exceeds the capillary force and the droplet becomes unstable. The disintegration of a charged droplet was pioneered by Zeleny [2], where the disintegration of a liquid jet issuing from an electrified capillary was achieved by applying an adequately high external electric field. For the first time, Macky [3] investigated the charged water droplet breakup phenomenon in the presence of a strong electric field. They reported that at a critical
applied field, the droplet elongates and filaments are drawn out from the ends due to surface instability. Photographic evidence of the droplet surface deformation, breakup, and jet formation was also reported. Further, the work of Macky [3] was supported by Taylor [4] using a hydrodynamic theory.

However, the study of an isolated charged droplet was systematically done by Doyle et al. [5], where, a Millikan oil-drop experiment setup [6] was used for droplet levitation. They observed that as the size of the droplet decreases the electric stress on the surface of the droplet increases due to inherent charge. Finally, the droplet ejects 1-10 smaller, highly charged progeny droplets along with 30% of its total charge. Like Doyle et al. [5], Abbas and Latham [7] reported similar results for larger sized droplet. Gomez and Tang [8] shown sub-Rayleigh (70 and 80% of the Rayleigh limit) breakup of free-falling heptane droplets where electrostatic sprays charge the droplets. Although numerous experimental studies have reported the critical limit of charge on the drop for the onset of Coulombic fission, the results of these studies show discrepancies. Duft et al. [9] provided unambiguous experimental confirmation of the Rayleigh limit of charge. The fission process remains unpredictable, and currently, no theory can accurately predict these fission-related losses. The reported values of charge and mass losses observed during Coulombic fissions vary from 10% to over 70%, and 0.1% to 30%, respectively [5, 7, 10, 11, 8, 12, 13, 14, 15, 16, 17, 11].

Theoretical analyses and numerical simulations have shown that, when the charge on the drop is equal to the Rayleigh limit, an initially perturbed spherical charged drop develops conical tips at the poles, and finally, a thin filament-like jet emerges from the tips (∼ ref [18, 19]) which confirm the experimental photographs of Duft et al. [9]. Very recently, Singh et al. [20, 21] have shown the frame-wise details of the droplet deformation, breakup of the droplet by jet detachment, and relaxation after the breakup. Although not accurate but the estimate of the charge and mass loss with the jets before the drop relaxes back to the stable spherical shape are also reported. Similar to Singh et al. [20], in the present work, we have reported high-speed imaging of levitated charged droplet and observed the entirely distinct mode of droplet breakup which is termed as an end-pinchoff mode of a breakup. Recently, Gañán-Calvo et al. [22] reported a similar mode of jet breakup in the electrospray setup and the droplet was found to be ejecting 29 to 40% of Rayleigh charge and 3% of its original mass.

Unlike Singh et al. [20] and Duft et al. [9], in the present work, the droplet is observed to eject 2 to 3 progeny droplets, and after the ejection of droplets, the jet relaxes back. The back-of-the-envelope calculations based on the theoretical scaling laws are derived for the number of progenies and jet diameter to supplement the experimental results. Although the droplet breakup and progeny formation depend on several parameters, as shown by Gañán-Calvo et al. [22], the theoretical analysis is carried out based on
initially known quantities such as the fraction of charge loss and mass loss. A remarkable agreement is observed between experiments and scaling laws.

2. Materials and method

The experiments described in the present work involve the levitation of ethylene glycol (EG) and ethanol mixture of a charged droplet. The appropriate amount of NaCl is added to vary the electrical conductivity of the droplet and measured using conductivity meter (Hanna instruments, HI 2316). The viscosity of ethylene glycol (EG) and ethanol mixture (50% v/v) droplet is measured using as Ostwald’s viscometer and found to be 0.006 Ns/m². The surface tension of the droplet is measured using the pendant drop (DIGIDROP, model DS) method and spinning drop (dataphysics, SVT 20 ) method and is found to be 0.03-0.04 N/m. The experiments are carried out at normal atmospheric conditions (1 atm pressure and 25 °C temperature).

In the present work, a positively charged droplet is levitated in a modified Paul trap. The detailed description of the setup can be found elsewhere [23]. The trap consists of two endcap electrodes and a ring electrode. The highlight of our trap is the significant value of $z_0$ ($\sim 6$ mm, distance between the centre of the ring to the bottom centre of the end cap electrode) and $r_0$ ($\sim 6$ mm, distance between centre of the ring to the inner periphery of the ring electrode) which provides enough space to perform several activities simultaneously such as introducing charged droplets generated by electrosprays in dripping mode, illuminating the drop and recording the drop deformation followed by Rayleigh breakup using high-speed camera (by Phantom V12 camera) at 1-1.3 hundred thousand fps. Both the endcap electrodes are shorted, and 11 kV$_{pp}$ voltage is applied using a high voltage amplifier (trek 8080) which is connected with a function generator for generating the desired waveform. The frequency is varied from 100-500 Hz for stable levitation of the droplet.

3. Experimental observations

In a typical experiment, the levitated sub-Rayleigh charged droplet undergoes evaporation and builds surface charge density with time. When the surface charge density exceeds beyond a critical limit, the droplet becomes unstable, deforms progressively to form jets and eventually breaks. The typical droplet deformation, jet formation, and breakup are shown in figure 1. It can be observed from the figure that the droplet breaks in the upward direction with an up-down asymmetry. Since the droplet is levitated in the presence of pure AC quadrupole field without superimposing any additional DC bias voltage to balance the gravity the droplet levitates slightly away from the geometric center of the trap. When a droplet is
levitated slightly away from the center of the trap it experiences a uniform field \( E = 4\Lambda z_{\text{shift}} \), where \( \Lambda \) is the intensity of the quadrupole field, \( z_{\text{shift}} \) is the z-directional downward distance from the center of the trap) which causes different electrical stress in the north pole and south pole of the drop. The droplet, therefore, breaks asymmetrically. In the first frame of the figure \( 1 \) a droplet is observed to form pear shape which suggests that the shape consists of a high magnitude of \( P_3 \) Legendre mode \(( \sim 0.056)\). The value of \( 3^{\text{rd}} \) Legendre coefficient \(( P_3 \) is obtained by fitting the outline of the experimental drop shape with the nonlinear least-square fitting method using software Mathematica\textsuperscript{®} (version 10). The drop shape outline is obtained using image processing software ImageJ (https://imagej.nih.gov/ij). When a charged droplet continues to evaporate the surface charge density reaches near Rayleigh charge. The high charge density with high initial positive \( P_3 \) perturbation results in instability that causes the droplet to break in the upward direction (see ref Singh et al. [20] for detailed explanation). In the present experiments, the jet is observed to break in an end pinch-off manner and relaxes back after ejecting a fraction of charge with a few countable numbers of progenies. We have conducted numerous experiments, and the end pinch-off phenomenon is observed only in 10% of the experiments. The specific circumstances for reproducibility of observation are not classified. However, a similar kind of breakup mode is observed in an electrospray setup of our laboratory, as shown in figure \( 2 \) as well as in the literature \(( \sim \text{see ref }[22])\). The breakup, maybe a chance encounter event, is observed in the downstream of electrospray with a high-speed camera at 1 hundred thousand fps frame rate.

Recently, Singh et al. [24] have reported several ways to measure the charge on the drop before and after the breakup. In the present experiments, we have employed the cut-off frequency method, and the obtained values are verified with the transient shift measurement method. The mass loss in the breakup process is obtained by measuring the size of the progenies using software ImageJ within \( \pm 6\% \) of the experimental error.

4. Problem formulation and validation

The droplet breakup is a result of an imbalance between the destabilizing electrical stress due to charge on the drop and the stabilizing capillary stress due to surface tension. At equilibrium, the electrical stresses which act normal to the surface of the drop equal the surface tension force. A droplet of radius \( R_0 \), as shown in figure \( 3 \) forms a jet during its breakup process and several progenies eject from the endpoints of the jet at the two poles of the drop. A schematic of droplet deformation and jet formation is shown in figure \( 3 \). We have considered the jet as a cylinder having a total surface charge \( Q \). The stress balance on the cylinder
can be written as,

\[ \frac{1}{2} \varepsilon_0 E^2 = \frac{\gamma}{a}, \]  

where, \( E = Q/(2\pi \varepsilon_0 a L) \), \( Q \) is the total charge on a cylinder of length \( L \), \( a \) is the radius of cylinder, as shown in figure 3. From the Gauss law, substituting the value of \( E \) in the equation 1, the resultant equation may be written as,

\[ \frac{Q^2}{8\pi^2 \varepsilon_0 L^2} = \gamma a. \]  

(2)

The radius of the cylinder in terms of known quantities (\( Q, L, \gamma \)) can be obtained by rearranging equation 2,

\[ a = \frac{Q^2}{L^2 \frac{1}{8\pi^2 \varepsilon_0 \gamma}}. \]  

(3)

Since it is difficult to measure the exact charge on the jet, we need to transform the equation 3 in terms of a fraction of charge loss and mass loss which is known to us. This can be achieved as,

\[ aL^2 = \frac{Q^2}{8\pi^2 \varepsilon_0 \gamma}. \]  

(4)

The volume of the cylinder is \( V = \pi a^2 L \) and corresponding \( a^2 L = V/\pi \) or \( a^4 L^2 = V^2/\pi^2 \). Further, dividing equation 4 by \( a^4 L^2 = V^2/\pi^2 \) results in the expression for the radius of the cylinder in terms of \( V \) and \( Q \), as given below,

\[ a^3 = (8\varepsilon_0 \gamma) \frac{V^2}{Q^2}. \]  

(5)
The quantities $V$ and $Q$ can be defined in terms of initially known quantities such as $V = f_v V_o$ and $Q = f_c Q_o$, where $f_v$ is the loosed volume fraction, $f_c$ is the loosed charge fraction. Thus, the radius of a cylinder given in equation (5) can be modified in terms of $f_v$ and $f_c$ by substituting the expression of $V$ and $Q$, given as

$$a^3 = 8\epsilon \epsilon_0 \gamma f_v^2 V_o^2 f_c^2 Q_o^2.$$  \hspace{1cm} (6)

Substitute, $Q_o^2 = 48\pi \epsilon \epsilon_0 \gamma V_0$, which is a critical charge required for droplet breakup, also known as Rayleigh charge, the resulting equation is shown below,

$$a^3 = \frac{f_v^2 V_0}{f_c^2 6\pi}.$$  \hspace{1cm} (7)
The above equation (7) can be transformed in terms of the radius of a sphere ($R_o$, shown in figure 3) which results in more convenient expression in the form of initially known parameters,

$$ a^3 = \frac{2 f_v^2}{9 f_c^2} R_o^3 . \quad (8) $$

Let us rewrite the radius $a$ in terms of $R_{jet}$ for better understanding. Upon performing algebraic manipulations equation (8) can be transformed as,

$$ a = R_{jet} = R_0 \left[ \frac{2 f_v^2}{9 f_c^2} \right]^{1/3} \quad (9) $$

Thus, equation (9) can be used for experimental comparison. Further, substituting the experimentally measured quantities, $f_v$ ($\sim 0.03$), $f_c$ ($\sim 0.31$) and $R_0$ ($\sim 80 \times 10^{-6}$) in equation (9) value of jet diameter ($d_{jet}$) can be obtained as,

$$ d_{jet} = 2 \times 80 \times 10^{-6} \left( \frac{2 \times 0.03^2}{9 \times 0.31^2} \right)^{1/3} = 21 \mu m. \quad (10) $$

Experimentally, the jet diameter is measured using ImageJ software. The experimentally obtained value of $d_{jet}$ and scale used for measurement is shown in figure 4. It can be observed from figure 4 that the value $d_{jet}$ is $\sim 23 \mu m$, which is in reasonable agreement with theoretically obtained value, i.e., $\sim 21 \mu m$.

The approach can be continued to estimate the number of progenies expelled during the droplet breakup process. Consider, the parent drop initially carries $Q_1$ charge and $V_1$ volume, and the residual charge and mass on the parent drop is $q_1$ and $v_1$. The total charge and mass can be given as, $\{Q_1, V_1\} \rightarrow \{nq_1, nv_1\}$,
where \( q_1 = \sqrt{48\pi\epsilon\epsilon_0\gamma v_1} = \sqrt{k v_1} \), \( k = \sqrt{48\pi\epsilon\epsilon_0\gamma} \), \( n \) is the number of progenies, \( q_1 = Q_1 / n \), \( v_1 = V_1 / n \), \( \frac{Q_1^2}{k} = k V_1 / n \), thus,

\[
    n = \frac{Q_1^2}{k V_1}.
\]

Similar to previous exercise the above equation can be transformed in terms of known variable \((f_v, f_c)\) and given as,

\[
    n = \frac{Q_1^2}{k V_1} = \frac{f_c^2 Q_0^2}{k_f V_0}. \tag{11}
\]

Form the Rayleigh limit we know that, \( Q_0^2 = k V_0 \). Hence the final expression for the number of progenies can be given as,

\[
    n = \frac{f_c^2}{f_v}. \tag{12}
\]

The equation 12 can be used for estimation of the number of progenies ejected during droplet breakup. Thus, for \( f_v \sim 0.03 \) and \( f_c \sim 0.31 \), \( n \) can be obtained as,

\[
    n = \frac{0.0961}{0.03} \approx 3
\]

From figure it can be observed that, during the breakup process exactly 3 progenies are ejected from the end point of the jet before it relaxes back. This validates the calculated value obtained from the equation 12. The following approach can obtain a similar expression.

To estimate the number of progenies, assume that the charge and the volume on the parent drop after the first ejection are \( Q_1 \) and \( V_1 \) respectively, where, \( Q_1 = Q_0(1 - f_c) \) and \( V_1 = V_0(1 - f_v) \), \( Q_0 \) and \( V_0 \) are the charge and volume of parent drop, respectively. The parent drop breaks into \( n \) offsprings, having charge \( q_1 \) and volume \( v_1 \),

\[
    Q_1 = (1 - f_c)Q_0 \Rightarrow Q_1^2 = (1 - f_c)^2Q_0^2 = (1 - f_c)^2k V_0,
\]

\[
    \frac{Q_1^2}{V_1} = \frac{(1 - f_c)^2k V_0}{V_0(1 - f_v)} = \frac{(1 - f_c)^2k}{(1 - f_v)}.
\]

Considering that the ejected daughter drops will carry same charge and volume, the remaining charge on the parent droplet is given as \( q_1 = f_c(1 - f_c)Q_0 / n \), similarly volume of off spring is \( v_1 = f_v(1 - f_v)^2Q_0^2 / (nk) \). We have seen in the previous section that,

\[
    q_1^2 = k v_1,
\]
substitute the functional form of $q_1$ and $v_1$ in above expression,

$$f_c^2(1-f_c)^2Q_0^2 = k \frac{f_v(1-f_v)^2Q_0^2}{nk}.$$  

Upon performing algebraic manipulations result in the number of off-springs ejected from the daughter drop,

$$n = \frac{f_c^2}{f_v}.$$  

Thus, we obtained the same expression as given in equation (12).

Since in the present experiments we observed primary (i.e., first breakup) breakup of a levitated charge droplet another interesting question that can be asked here is what the extent of ejection from the parent drop is? If we consider that the droplet ejects the same number of progenies in each successive breakup. It is an important exercise to calculate the extent of mass loss during the lifetime of a charged droplet. The droplet loses its mass in two ways first is the loss due to breakup of droplet and second is the mass due to evaporation, i.e., total mass lost= mass lost due to evaporation + mass loss due to rayleigh breakup. 

The schematic representation of total mass loss is shown in the figure 5. The Horizontal movement in the figure represents the charge and mass loss due to Rayleigh breakup while the vertical downward movement shows the mass loss due to evaporation. The mass loss in each successive breakup is $\sim f_v Q_o^2$ while the remaining mass is $\sim (1-f_v)Q_o^2 = g_v Q_o^2 = g_c Q_o^2$. Hence, the total mass loss can be calculated by referring figure 5 i.e.,

$$\text{mass lost} = f_v + f_c g_v^2 + f_v g_c^4 + f_c g_c^6 + f_c g_c^8.$$  

The equation (14) is the simple geometric progression of $g_c$. Thus, expression for the total mass loss can be obtained as,

$$\text{mass lost} = \frac{f_v}{1-g_c^2}.$$  

For, $f_v=0.03$, $g_c=1-f_c=1-0.31=0.69$ and $g_v=1-f_v=1-0.03=0.97$, the mass lost is $0.03/(1-0.69^2)=0.047 \sim 5.8\%$

In the droplet breakup process, one can ask another interesting question which is what will be the life span of a charged droplet? For example, the mass loss in the droplet breakup process takes place via two different ways, Rayleigh breakup and evaporation, before it completes its life time. The time taken by a droplet during evaporation is longer than that of the Rayleigh breakup time. Hence first the time taken by the evaporation is calculated and then the correction factor for accounting the total time of breakup is multiplied. The total life time of the drop can be calculated by diagrammatic model that has been developed
Figure 5: The flow of charge loss and mass loss of parent droplet during evaporation or breakup

in figure 5 to evolve the generational history of drop. The rate of change of size due to evaporation can be given as,

\[ t_1 = \frac{Q_0^{2/3}}{K}(g_v^{2/3} - (g_c^{2/3})) \]  

(16)

t_1 is the time required for the mass loss due to evaporation after 1st breakup of droplet, \( Q_0 \) is the initial charge on the droplet, \( K \) is the evaporation rate constant, \( g_v, g_c \) has usual definitions. Similarly,

\[ t_2 = \frac{Q_0^{4/3}}{K}((g_v g_c^{2/3} - (g_c^4)^{2/3}), \]  

(17)

\[ t_3 = \frac{Q_0^{4/3}}{K}((g_v g_c^{4/3} - (g_c^6)^{2/3}), \]  

(18)

\[ t_4 = \frac{Q_0^{4/3}}{K}((g_v g_c^{6/3} - (g_c^8)^{2/3}), \]  

(19)
and can be generalized for \( n \text{th} \) order which is given below,

\[
t_n = \frac{Q_0^{4/3}}{K} ((g_v c_v^{2(n-1)})^{2/3} - (g_v c_v^{2n})^{2/3}),
\]

(20)

further simplification can result in the following form,

\[
t_n = \frac{Q_0^{4/3}}{K} \left[ g_v^{4n/3} \right] \left[ g_v^{2/3} - \left( \frac{g_v^{2n}}{g_v^{4/3}} \right) \right].
\]

The above expression can be represented in the more simpler form as, \( t_n = A \phi^n \), where,

\[
A = \frac{Q_0^{4/3}}{K} \left[ g_v^{4/3} \right] \left[ \frac{g_v^{2/3}}{g_v^{4/3} - 1} \right].
\]

The total time is the sum of \( t_1, t_2, ..., t_n \), which is given as,

\[
\sum t_n = A \sum_{n=1}^{\infty} \phi^n - 1
\]

\[
= A \left[ \frac{1}{1 - \phi} - 1 \right]
\]

Substituting the \( A \) and \( \phi \) in the above expression results in

\[
t = \frac{Q_0^{4/3}}{K} \left[ g_v^{4/3} - \left( \frac{g_v^{2n}}{g_v^{4/3}} \right) \right].
\]

(21)

It can be noticed in the equation 21 that all quantities are known except \( k \) which is the diffusion coefficient.

By simple theory of diffusion and neglecting temperature change, the rate of change of volume can be given by,

\[
V_i^{2/3} - V_j^{2/3} = \left[ \pi \left( \frac{a}{2} \right)^2 \right]^{2/3} 8D_v c_s v_m t_i = k_D t_i.
\]

(22)

Where, \( k_D = \left[ \frac{\pi}{6} \right]^{2/3} 8D_v c_s v_m \), \( v_m = m/\rho = M/(N_A \rho L) \), \( c_s = p_s/(k_B T) = p_{eq}/(k_B T) \), \( p_{eq} \) is equilibrium partial pressure of droplet, \( D_v \) is diffusivity of the droplet, \( k_B \) is the Boltzmann constant, \( T \) is the temperature, \( p_s \) and \( p_{eq} \) are the saturation and equilibrium pressure respectively and \( \rho_L \) is the density of the liquid.

For the sake of clarification let us solve equation 21 by substituting values for ethylene glycol droplet.

\( f_v = 0.03, \ f_c = 0.3, \ g_v = 1 - f_v, \ g_c = 1 - f_c, \ a = 80 \times 10^{-6} \ \mu m, \ k_B = 1.381 \times 10^{-23}, \ T = 298.0 \ K, \ N_A = 6.02 \times 10^{23}, \ D_v = (0.108 \times 10^{-4})(/293)^{1.75}, \ M = 62.07 \times 10^{-3} \ Kg, \ \rho = 1113.0 \ Kg/m^3, \ AA = 8.7945, \ Bb = 2615.4, \ Cc = 244.91, \ p_{eq} = 132 \times 10^{(AA - Bb/(Cc + t - 273))} \). The time for evaporation is calculated as

\[
t_{\text{evaporation}} = V_i^{2/3}/k_D = \frac{1.66 \times 10^{-8}}{2.18573 \times 10^{-11}} = 761 \ \mu s
\]
It can be observed that the magnitude of evaporation time is very less this is due to parameters which are taken for calculation of diffusivity may not correspond to the present situation. Further the time taken in the Rayleigh breakup process can be obtained form the following expression,

\[
t_{\text{Ray}} = t_{\text{evaporation}} \left[ \frac{g_{v}^{4/3} - (g_{c}^{2})^{4/3}}{1 - (g_{c}^{2})^{4/3}} \right]^{1/2} = 761 \times \frac{[0.97^{4/3} - (0.69^{2})^{4/3}]}{1 - (0.69^{2})^{4/3}} = 713\mu s
\]  

It is interesting to note that the total time require to evaporation \( t_{\text{evaporation}} \) and the total time required for Rayleigh breakup \( t_{\text{Ray}} \) is almost same.

5. Conclusions

The high-speed imaging of the breakup process of a charged droplet levitated in an electrodynamic balance is reported in this work. To the best of our knowledge, this is the first study which presents the experimental observation of end pinching mode of breakup of a critically charged droplet. A similar mode of droplet breakup is also observed in the high-speed imaging of electrospray. The experiments indicate that a levitated charged droplet ejects three equal sized highly charged progeny droplets from the tips of the drop and the jet relaxes back after ejecting 31% charge, and about 3% mass of the original droplet. This contradicts the previously reported studies where the jet detachment is observed during the breakup process of an isolated charged drop [9]. Based on the charge and mass loss information from the experiments, simple calculations are carried out to predict the jet diameter, number of progeny droplets and the life span of a charged droplet. The calculations are in fair agreement with the experimental observations.

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