Experimental Study on Non-coalescence Characteristics of Oppositely Charged Droplets (Postprint)

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Abstract

Based on microscopic high-speed digital imaging technology, an experimental study was conducted on the evolution process of oppositely charged droplet collision behavior. Using hydrochloric acid aqueous solutions with different ionic concentrations as the research medium, the morphological characteristics of non-coalescence and fragmentation of oppositely charged droplets under different voltages were precisely captured, and the influence patterns of medium conductivity on the non-coalescence and fragmentation characteristics were investigated. The experimental results indicate that: for low-conductivity droplets, the dynamic behavior under higher voltages manifests as non-contact rebound. With increasing conductivity, the dynamic behavior of the droplet collision process manifests as non-contact fragmentation, where the ion concentration at the droplet tip reaches the Rayleigh limit before air discharge occurs. The intensity of droplet fragmentation behavior is proportional to the liquid conductivity; for high-conductivity liquids, droplet fragmentation behavior is observed at lower voltage ranges, and the droplet stretching rate and fragmentation volume are more sensitive to changes in electric field strength.

Full Text

Experimental Study on Non-coalescence Characteristics of Oppositely Charged Droplets

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Abstract

Based on high-speed microscopic digital imaging technology, an experimental investigation was conducted on the evolution of collision behavior between oppositely charged droplets. Using aqueous hydrochloric acid solutions with different ion concentrations as the test medium, the non-coalescence and breakup morphological characteristics of oppositely charged droplets under various voltages were accurately captured, and the influence of medium conductivity on the non-coalescence and breakup behavior was explored. Experimental results demonstrate that the dynamic behavior of low-conductivity droplets at higher voltages manifests as non-contact bouncing. As conductivity increases, the dynamic behavior during droplet collision transitions to non-contact breakup, where the ion concentration at droplet tips reaches the Rayleigh limit before air discharge occurs. The intensity of droplet breakup is directly proportional to liquid conductivity; high-conductivity liquids exhibit droplet breakup at lower voltage ranges, and both the droplet stretching rate and breakup volume become more sensitive to changes in electric field strength.

Keywords: oppositely charged droplets, ion migration, Coulomb fission, microscopic high-speed digital imaging

Introduction

Charged droplets and their electric field-induced dynamic behaviors constitute fundamental aspects of electrohydrodynamics. Technologies based on charged droplets, such as electrostatic atomization, have found widespread applications in spray coating, thin film preparation, air purification, space micro-propulsion, and biological mass spectrometry. The electrostatic atomization process involves complex coupling mechanisms between droplet coalescence and separation, with Coulomb forces and surface tension interacting in intricate ways, making the dynamic behavior between adjacent charged droplets not yet fully understood. While minimum surface energy theory suggests that adjacent droplets will fuse upon contact, the contact behavior between neighboring oppositely charged droplets is particularly unique, with novel phenomena such as non-coalescence and breakup attracting significant research interest from scholars worldwide, and recent progress has been made in fundamental studies [1,2].

Ristenpart et al. [3] demonstrated that when droplet charge reaches a certain level, oppositely charged droplets exhibit contact bouncing during collision, attributing this behavior to changes in droplet geometry depending on the relationship between curvature at the liquid bridge and local capillary pressure. Bird et al. [4] proposed a surface energy model to describe the transition from coalescence to bouncing in oppositely charged droplets. Helmensdorfer [5] developed a new model based on mean curvature flow to describe this transition, with the advantage of eliminating the need for precise measurement of liquid bridge geometry. In summary, considerable detailed research has been conducted on the contact bouncing behavior between oppositely charged droplets. However, when liquid conductivity reaches certain levels, new dynamic behaviors such as non-contact breakup between adjacent droplets have rarely been reported, and both non-contact bouncing and contact breakup lack reasonable explanations. Consequently, relevant research is urgently needed to investigate the non-coalescence and breakup mechanisms of oppositely charged droplets.

This study employs high-speed digital imaging technology with high spatiotemporal resolution to investigate the non-coalescence and breakup characteristics of oppositely charged droplets under electric fields. From the perspective of ion migration and based on Rayleigh limit theory, the Coulomb fission behavior of oppositely charged droplets is analyzed. Breakup volume and stretching rate are defined to describe breakup intensity, aiming to reveal the non-coalescence and breakup mechanisms between adjacent droplets through accurate capture and post-processing analysis of morphological features during droplet collision.

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1 Experimental Setup and Methods

[Figure 1: see original paper] shows the experimental apparatus for oppositely charged droplet collision, constructed entirely of organic glass. The downstream droplet positioned on a superhydrophobic substrate and the upstream droplet suspended from a copper wire are strictly symmetrical. A negative high-voltage electrostatic generator (0–20 kV) connects its high-voltage terminal to the upstream copper wire, while the downstream superhydrophobic substrate is grounded, enabling the upstream and downstream droplets to acquire opposite charges. Subsequently, the upstream droplet is slowly moved vertically downward toward the downstream droplet using a micrometer adjustment.

An Olympus i-speed3 high-speed digital camera equipped with a microscopic zoom lens (for magnifying the camera' s field of view) captures the experiments. The system records the evolution of droplet collision morphologies at 15,000 fps for different conductivity liquids under varying voltages, enabling post-processing analysis. The light source is a 40 W high-brightness LED cold light positioned opposite the high-speed camera and directly facing the oppositely charged droplets, ensuring clear images even at extremely short exposure times. The image resolution is 420×312 pixels.

Deionized water was used to dilute 6 mol/L hydrochloric acid to obtain aqueous hydrogen chloride solutions of different mass fractions. The conductivity of each solution was measured using a conductivity meter, as shown in Table 1. Except for conductivity (which varies by three orders of magnitude), the physical properties of these solutions—such as viscosity and surface tension—show minimal variation, allowing investigation of conductivity' s influence on the dynamic evolution of oppositely charged droplets. The experimental environment temperature was maintained at 25±0.5°C.

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2.1 Charge Distribution on Charged Droplets

When a stationary droplet acquires net charge, Coulomb repulsion distributes the charge on the droplet surface [6]. Theoretically, the surface charge density is uniform, and droplet stability depends on the balance between surface tension pressure (P__s) and expansion pressure (P__e) caused by surface tension and Coulomb repulsion, respectively, as shown in Figure 2 [Figure 2: see original paper]. Early analysis of charged droplet breakup suggested that when surface charge density reaches a certain level, droplets become unstable and break up. This critical limit is known as the Rayleigh limit [7], where is the dielectric constant of ambient air, is droplet surface tension, and r is droplet radius. Q represents the critical charge at which P__s = P__e. The Rayleigh limit can be explained by analyzing the relationship between surface energy (4 r^2) caused by surface tension and electrostatic energy ($Q^2/(8\ r)$) caused by Coulomb repulsion. Surface energy maintains droplet stability and prevents deformation, while electrostatic energy enhances instability. Based on the principle of virtual work, the difference in work done by these forces reflects the droplet' s stability. When $\Delta W = 0$, the work done by both forces is equal. Typically, surface charge density describes the charging state, and the critical charge density can be derived from the Rayleigh limit. This parameter indicates that breakup occurs when surface charge density reaches a certain level, enabling description of droplet instability through ion concentration differences caused by ion migration.

However, theoretical analyses of charged droplet stability are based on the assumption of uniform surface charge distribution, where droplet breakup occurs when overall surface charge density reaches the Rayleigh limit. In reality, under electric fields, surface charge distribution on charged droplets is non-uniform, with charges migrating along the electric field direction to localized tip regions, as shown in Figure 3 [Figure 3: see original paper]. If the electric field intensity is sufficiently high, droplet surface charge distribution becomes extremely non-uniform, causing localized charge accumulation to first reach the Rayleigh limit and triggering localized Coulomb fission. At this point, the droplet' s total charge has not yet reached the Rayleigh limit, meaning the droplet itself undergoes localized instability breakup when reaching a certain sub-Rayleigh limit [9].

2.2 Evolution Behavior of Oppositely Charged Droplet Collisions Under Conductivity Influence

High-conductivity droplets contain high ion concentrations, leading to greater charge accumulation at localized tip regions under electric fields and making it easier for local charge density to reach the Rayleigh limit and cause instability breakup. Figure 4 [Figure 4: see original paper] shows the dynamic evolution morphologies of oppositely charged droplets at 820 S/cm conductivity. As voltage increases, two new dynamic behaviors emerge: contact bouncing and non-contact breakup. At 1.0 kV, the behavior transitions from coalescence to contact bouncing, which can be reasonably explained by ion migration mechanisms from physical chemistry [10]. When oppositely charged droplets contact, a liquid bridge forms, allowing negative ions to migrate from the negative to positive electrode through the bridge while positive ions migrate in the opposite direction. Each droplet tip instantaneously accumulates like charges migrating from the other tip, creating instantaneous Coulomb repulsion at both tips. Once the initial surface charge density at droplet tips reaches a certain level, this instantaneous Coulomb repulsion dominates the bouncing behavior. Therefore, judging droplet coalescence behavior solely through tip curvature and local capillary pressure relationships—without considering instantaneous Coulomb repulsion—is inaccurate, unless assuming sufficiently low ion concentrations to neglect this effect. In low-conductivity cases, fewer ions result in weaker instantaneous Coulomb repulsion that cannot induce bouncing during contact.

At 1.2 kV, non-contact breakup occurs, a special behavior that only appears when droplet conductivity reaches a certain level. As oppositely charged droplets gradually approach, electric field intensity at droplet tips increases, causing more charge accumulation and increasing charge density to the Rayleigh limit, triggering tip Coulomb fission. This means non-contact breakup occurs when droplet tip charge density reaches the Rayleigh limit before air discharge. Breakup releases substantial charge at droplet tips, instantly reducing electrostatic attraction between droplets, after which surface tension causes them to rebound. This approach-breakup-rebound cycle repeats. As voltage further increases, ion migration accelerates, enlarging the region at droplet tips that reaches the Rayleigh limit within the same timeframe, increasing breakup volume and intensity, as shown in Figure 4(4).

The relationship between liquid conductivity and charging voltage determines charge density at droplet tips, with secondary breakup occurring only when the Rayleigh limit is reached. Figure 5 [Figure 5: see original paper] shows the critical charging voltage for droplet breakup as a function of conductivity. Curve fitting using Origin 8.0 reveals that each conductivity has a minimum critical voltage inducing secondary breakup. Under current experimental conditions, this boundary line distinguishing non-contact bouncing from non-contact breakup can predict droplet dynamic behavior for any conductivity-voltage combination. High-conductivity liquids require lower critical voltages for breakup, while low-conductivity liquids need higher voltages. Deionized water with 1.3

S/cm conductivity showed no breakup behavior under current experimental conditions, indicating that conductivity plays a crucial role in the breakup behavior of oppositely charged droplets.

To quantify breakup intensity, breakup volume V_b is defined as the volume ejected from droplet tips 2/15 ms after breakup initiation. Breakup volume is determined by measuring the cone base radius and height. This quantification enables more intuitive understanding of breakup intensity. Figure 6 [Figure 6: see original paper] shows breakup volume as a function of voltage for different conductivity liquids. Breakup volume increases with voltage for all conductivities because higher electric field intensity accelerates ion accumulation at droplet tips, expanding the breakup region that reaches the Rayleigh limit. However, growth rates differ significantly; at the same voltage, high-conductivity droplets exhibit larger breakup volume due to greater instantaneous ion migration, indicating that high-conductivity droplets are more sensitive to electric field intensity changes.

Conclusions

This experimental study investigated the non-coalescence and breakup behavior of oppositely charged droplets under conductivity influence, yielding the following conclusions:

(1) In the low-conductivity regime, increasing voltage first produces coalescence followed by non-contact bouncing behavior. Droplet tip charge density always undergoes air discharge before reaching the Rayleigh limit, and no Coulomb fission is observed.

(2) In the high-conductivity regime, increasing voltage produces contact bouncing followed by non-contact breakup behavior. The instantaneous Coulomb repulsion at liquid bridge ends caused by ion migration plays a crucial role in bouncing behavior. Non-contact breakup indicates that droplet tip charge density reaches the Rayleigh limit before air discharge occurs.

(3) The fitted voltage-conductivity curve shows that breakup becomes more likely as conductivity increases, while insulating liquids with extremely low ion concentrations cannot induce breakup behavior. Increased conductivity enhances droplet sensitivity to electric field intensity changes, enlarging breakup volume and accelerating stretching rate.

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Note: Figure translations are in progress. See original paper for figures.