Astonishing life of a coalescing drop on a free surface

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We report an interesting feature in the consecutive steps of coalescing of a drop, which is called a cascade of partial coalescence. It is observed that as the secondary drop gets smaller, it bounces higher. We show that the capillary force is the main driving force for this phenomenon. By using ultra-high-speed video, it is revealed that the capillary force at the pinch off pulls the drop to the planar interface. The drop bounces off the interface and moves upward until it reaches the maximum height. A theory is developed that includes the capillary and gravitational forces and predicts this process.

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Every time a raindrop hits a puddle, a wave breaks, you pour milk into coffee, as simple as it may look, there are phenomena that are unseen to the naked eye. The coalescence of a liquid drop with a planar layer of the same liquid has been the subject of research since the 19th century. Rayleigh [1,2], Thomson and Newall [3], and Reynolds [4,5] reported several features of this phenomenon. When a liquid drop approaches a planar interface of the same liquid and air, the result of impact can be a complete engulfment of the drop in the pool of liquid (splashing) [8–18], a rebound off the free surface (bouncing or skipping) [7,19], or a temporary noncoalescence situation which is due to the existence of a thin layer of air between the drop and the planar liquid. After a finite time, the air will be drained out and this layer will be thin enough for coalescence to occur [3,6,7,11,13,20–28]. It has been shown that the drop size R, surface tension of liquid σ, viscosity μ, density ρ, and velocity (and angle) of impact V_i are important parameters for this phenomenon. The dimensionless numbers of this problem are the Ohnesorge number $\mathrm{Oh}=\mu/\sqrt{\rho R\sigma}$, the Weber number $\mathrm{We}=\rho V_i^2 R/\sigma$, and the Froude number $\mathrm{Fr}=V_i^2/2gR$. Depending on the values of these dimensionless numbers, the drop splashes, bounces, or coalesces. The condition for each regime have been fully documented in the literature [11,13,16,26]. If a coalescing drop ($\mathrm{We}\sim 1$) has physical properties such that the Ohnesorge number is smaller than unity, it has been shown that instead of a complete engulfment, a secondary drop or drops will form as a result of partial coalescence. The cascade of partial coalescence of the secondary drops continues until the radius of the last drop is small enough ($\mathrm{Oh}\sim 1$). At this point, the drop is completely engulfed into the planar liquid. This cascade has been observed and analyzed by Charles and Mason [22] (who described the mechanism of the process), and Thoroddsen and Takehara [6] (who described the time scale of the process).

A feature of this cascade, which has never been previously explained, is the bouncing of the secondary drops off the liquid interface. The maximum height after the bounce is typically higher for smaller drops. A schematic of formation of the secondary drop and the bouncing is shown in Fig. 1(a). The initial drop, with radius R_1, is deposited on the planar liquid/air interface by a syringe and needle system. The syringe was mounted in vertical position and approximately 4 mm from the fluid/air interface to not only minimize the impact velocity of the initial drop ($\mathrm{We}\sim 1$) but also allow the drop to fall clear of the needle before touching the surface. The container of fluid was constructed large enough to eliminate the effect of sidewalls on the experiment. When the initial drop reaches the interface, there is a thin layer of air between the drop and the liquid/air interface that drains due to the weight of the drop. When the initial drop finally contacts the interface, a hole is formed through which the fluid inside the drop moves downward. During this process vortex rings [29] in the bulk fluid and capillary waves on the interface are observed. This process leads to a pinch-off process [6,22] that generates the secondary drop. Before the pinch off, the fluid leaving the drop forms a cylindrical shape that later evolves into a secondary drop with radius of R_2. The height of this column is approximately $2R_1$ and its radius R_0 is assumed to be constant [22] as shown in Fig. 1. The secondary drop moves downward, bounces on the interface, and moves upward until reaching a maximum height h_{max}, before falling down back to the surface, coming to a rest, and becoming the primary drop for the second step of the cascade. These steps continue until the drop radius becomes too small and the Ohnesorge number becomes larger than unity and consequently no secondary drop is produced.

A series of frames in the coalescence cascade for a water drop is presented in Fig. 1(b). The reference time is measured from the first frame. The first drop in the series is 0.60 mm in radius; its coalescence occurs until 7.83 ms, at which time its secondary drop of radius 0.32 mm begins bouncing and attains a maximum height of 1.54 mm, measured from the water surface to the center of the drop, at a time 20.3 ms. The coalescence of this drop occurs from a time of 65.9 to 68.6 ms, at which time the next drop of radius 0.16 mm begins bouncing and attains a maximum height of 1.24 mm at a time of 84.8 ms from the first frame. A complete trajectory of this cascade is shown in Fig. 2. The vertical axis of this graph is the height in mm and the horizontal axis is time in seconds. The circles at the end of each trajectory are the schematics of the drops for each path with actual radii. As shown in Fig. 2, the maximum height after bounce, h_{max}, is lower for the second step compared to the first step and third steps [31]. This observation contradicts previous observations of the smaller drops always bouncing higher [6].

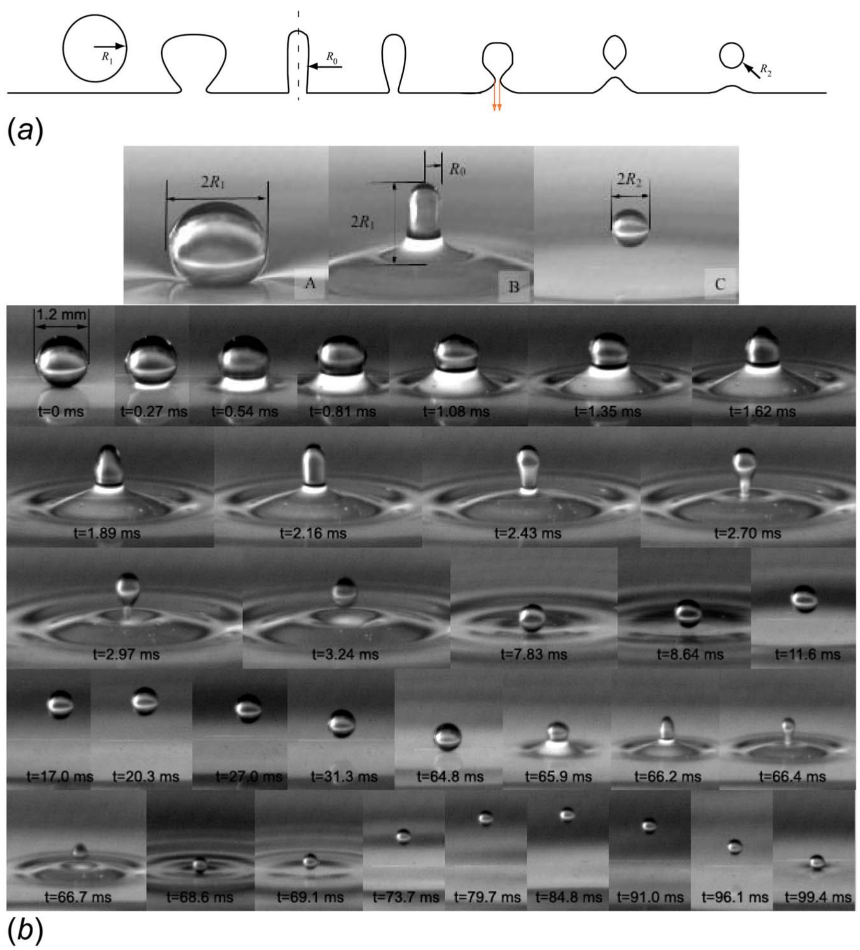

FIG. 1. (Color online) (a) Schematic of the partial coalescence of a drop, with radius R_1, into a pool of the same fluid. The secondary drop R_2 is formed from a column of liquid with radius R_0 and height of approximately $2R_1$, after the pinch-off process. (b) A series of frames in the coalescence cascade of a water drop. The time given in each frame is measured from the first frame. The liquid column, the pinch-off process, and bouncing of the secondary drops are shown in this set.

During the pinch off, there are two forces applied to the secondary drop: the gravitational force $mg=4/3\rho\pi R_2^3 g$, and the capillary force $2\pi R(t)\sigma$. The radius at the pinch-off cross section, $R(t)$, is initially R_0 and eventually becomes zero. The initial radius R_0 can be found by equating the volume of the column with the secondary drop [22], which gives $R_0=\sqrt{2/3r_i^3}R_1$ where $r_i=R_2/R_1$ is the drop ratio. Newton's second law requires that $(4/3)\rho\pi R_2^3 g+2\pi R(t)\sigma=(4/3)\rho\pi R_2^3 dV/dt$. By integrating in time from zero to $\tau=\sqrt{\rho R_1^3/\sigma}$, which is known to be the time scale of this problem [6], and in velocity from zero to the initial downward velocity of the secondary drop, V_0, a relation for initial velocity can be derived. It is known that the radius at the pinch-off cross section varies with time [30] as $R(t)=R_0(1-e^{(t-\tau)/\tau})$. Thus the initial downward velocity of the secondary drop becomes $V_0=e^{-1}\sqrt{3\sigma/2\rho r_i^3 R_1}+g\sqrt{\rho R_1^3/\sigma}$. The incident velocity of the drop when it impacts the interface is $V_i=\sqrt{4gR_1+V_0^2}$ assuming the secondary drop falls down from the height of $2R_1$. Also from kinematics, the rebound velocity is $V_b=\sqrt{2gh_{max}}$, assuming the viscous effect in air is negligible since the Stokes number $(\mathrm{St}=\mu V_b/\rho g R^2)$, which is the ratio of viscous force to gravitational force, is much

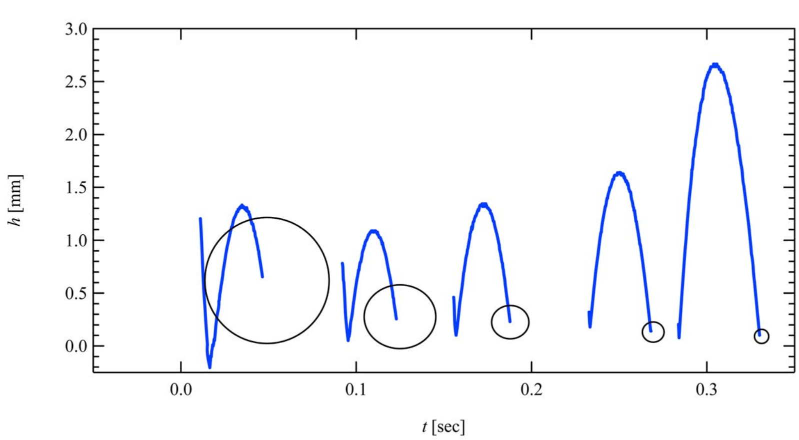

FIG. 2. (Color online) The trajectory of secondary drops bouncing off the interface of liquid/air during the coalescence cascade. The circles represent the actual sizes of the secondary drops. The discontinuity in the trajectory is due to the residence time of each drop before the next stage of coalescence.

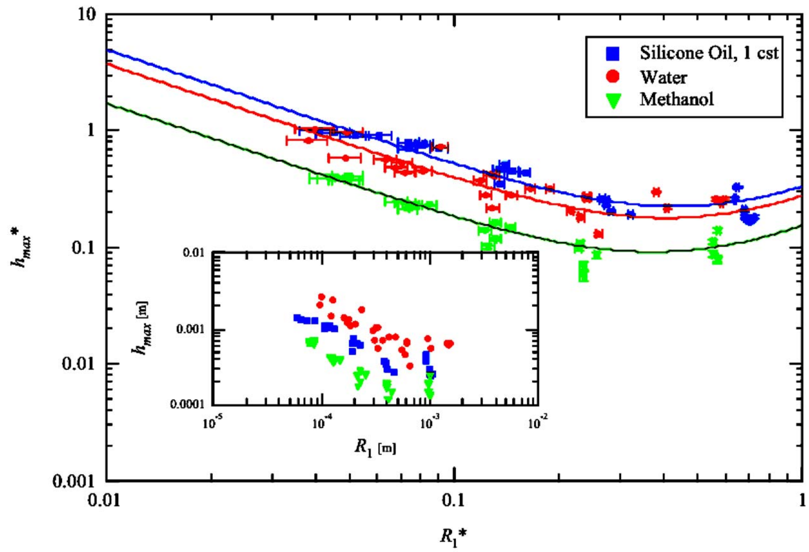

FIG. 3. (Color online) The relation between the dimensionless maximum height h^*_{max} and the dimensionless radius of the drop R^*_1 for methanol (triangle), water (circle), and silicone oil (square). The solids lines are the best fits of each set of data to $h^*_{max}/A^2 = 3/(4e^2 r_i^3 R^*_1) + R^*_1[2+\sqrt{3/(2e^2 r_i^3)}] + R_1^{*3}/2$ with A and r_i as the fitting parameters. As the radius of the drop increases, the maximum height decreases until it reaches a minimum value. For larger drops, the maximum height increases with the size of the drop due to the gravitational effect. Inset is the dimensional plot of the maximum height h_{max} in meters as a function of the radius R_1 in meters for the same data.

smaller than unity. The coefficient of restitution for the bouncing impact is defined as $A=V_b/V_i$. This leads to the relation between the dimensionless maximum height $h^*_{max} = h_{max}/l_{cap}$ and the dimensionless primary radius $R^*_1 = R_1/l_{cap}$, where $l_{cap}=\sqrt{\sigma/\rho g}$ is the capillary length of the fluid, as $h^*_{max}/A^2 = 3/(4e^2 r_i^3 R^*_1) + R^*_1[2+\sqrt{3/(2e^2 r_i^3)}] + R_1^{*3}/2$.

A series of experiments were conducted with water ($\sigma = 70\times 10^{-3}$ N/m, $\rho=1000$ kg/m^3, $\mu=1\times 10^{-3}$ Pa s), methanol ($\sigma=23\times 10^{-3}$ N/m, $\rho=792$ kg/m^3, $\mu=5.8\times 10^{-4}$ Pa s), and Dow® 200 silicone oil ($\sigma=17\times 10^{-3}$ N/m, $\rho=820$ kg/m^3, $\mu=8.2\times 10^{-4}$ Pa s) for validation of our hypothesis. The surface tensions and viscosities of these fluids were measured in our laboratory before each experiment. For a bouncing drop, h_{max} and R_1 were measured using high-speed video. The dimensionless maximum height h^*_{max} is plotted as a function of the dimensionless initial radius R^*_1 in Fig. 3. The dimensional form of this plot is also presented at the inset of Fig. 3. As the drop radius decreases, the maximum height decreases to a minimum value, and then increases in a power-law form. It is not surprising that h^*_{max} values for all materials are not completely collapsed on a

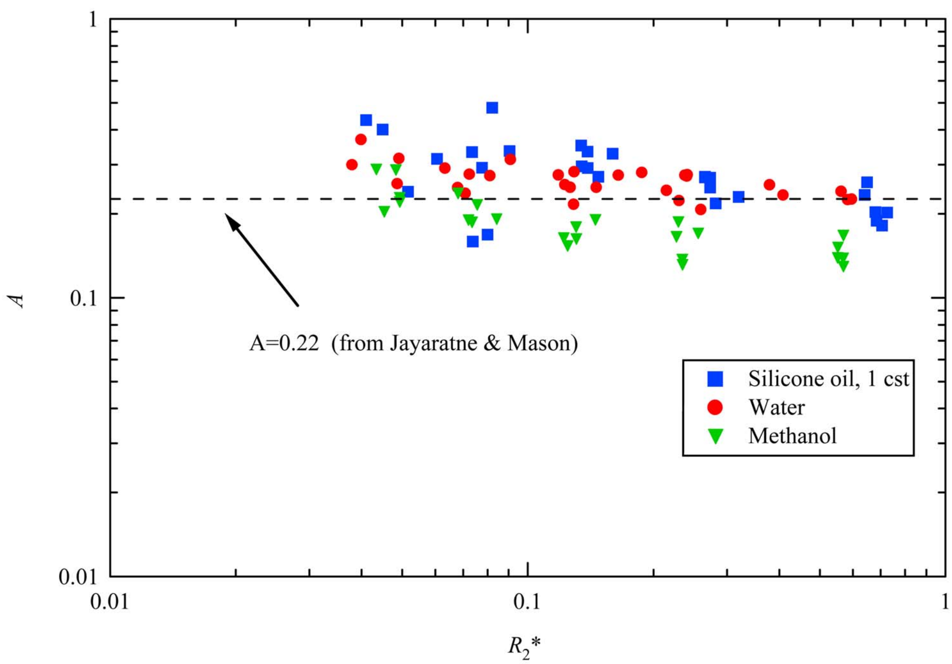

FIG. 4. (Color online) The relation between the coefficient of restitution A and the dimensionless radius of the bouncing drop R^*_2 for methanol (triangle), water (circle), and silicone oil (square). The dashed line is the value $A=0.22$ given by Jayaratne and Mason [7] for water, extrapolated from the data for skipping drops.

master curve. The generation of vortices and surface waves are among several sources of energy dissipation during the bouncing process. The coefficient of restitution A depends on the size of the drop and the physical properties of the surface; thus it is not the same for the three materials, which explains the shift in the dimensionless data. The solid lines are theoretical data from the theory for each material. Our theoretical fitting parameters for each material are found to be $A_{meth}=0.21\pm0.08$, $r_i=0.65\pm0.08$; $A_{water}=0.27\pm0.01$, $r_i=0.58\pm0.06$; and $A_{SiOil}=0.27\pm0.03$, $r_i=0.52\pm0.05$. We have also observed that the drop ratio r_i is not a constant value from the experiment. The drop ratio values are found to have a range of $0.5<r_i<0.7$. The curve fitting leads to an average r_i for each material that is within the range of observed values. We combined our predicted equation and the measured values of r_i, h_{max}, and R_1 to calculate the coefficient of restitution A. The values of A as a function of initial radius R_2 are presented in Fig. 4. As shown in this figure, the coefficient of restitution is approximately constant for all the values of R_2. As expected, the average value of the coefficient of restitution is different for each material. Methanol has the lowest coefficient of restitution of $A=0.20$, followed by water, $A=0.26$, and silicone oil, $A=0.28$. The only reported value for the coefficient of restitution is given by Jayaratne and Mason [7]. They measured the coefficient of restitution for series of water drops that bounce off the water surface when they impact with an angle less than 90°. The coefficient of restitution for normal impact was found by extrapolation of the data to be $A=0.22$, shown in Fig. 4 with a dashed line. This value agrees with our direct measurement of the coefficient of restitution for water.

In summary, we reported the phenomenon of the coalescence-induced bouncing of droplets at gas/liquid interfaces during the coalescence cascade of liquid drops. It is revealed that the capillary force at the pinch off pulls the drop to the planar interface. The drop then bounces off of the interface and moves upward until it reaches the maximum height. We showed that the capillary force is the main driving force for this phenomenon. We developed a theory that includes the capillary and gravitational forces and predicted the maximum height of the bouncing drop. We showed that this theory matches well with the experimental results for several fluids such as water, methanol, and silicone oil. We also measured directly the coefficient of restitution for the normal impact of a drop with the planar liquid/air interface. The measured values of the coefficient of restitution have good agreement with the theoretical predictions.

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[31] See EPAPS Document No. E-PLEEE8-73-097602 for a movie of the cascade process. This document can be reached via a direct link in the online article's HTML reference section or via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html).