The coalescence-cascade of a drop

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Abstract

When a drop is deposited gently onto the surface of a layer of the same liquid, it sits momentarily before coalescing into the bottom layer. High-speed video imaging reveals that the coalescence process is not instantaneous, but rather takes place in a cascade where each step generates a smaller drop. This cascade is self-similar and we have observed up to 6 steps. The time associated with each partial coalescence scales with the surface tension time-scale. The cascade will however not proceed ad infinitum due to viscous effects, as the Reynolds number of the process is proportional to square root of drop diameter. Viscous effects will therefore begin to be important for the very smallest drops. This cascade is very similar to the one observed previously by Charles & Mason [*J. Colloid Sci.* **15**, 236 (1960)] for two immiscible liquids, where one of the liquids replaces the air in our setup.

We report here a curious phenomenon, which occurs when one deposits a drop onto a layer of the same liquid. The drop hesitates briefly before coalescing into the bulk fluid, due to the draining of a thin layer of air sitting between the two liquid masses. As contact is established the unbalanced surface tension forces initiate a capillary wave which greatly deforms the drop which coalesces only partially, pinching off a new drop at its top, as shown in Fig. 1(a). The daughter drop bounces and comes to rest at the surface, repeating this partial coalescence. We have observed up to 6 steps in this cascade, starting with drop diameters around 3 mm and decreasing in diameter by approximately a half during each step.

Such cascades have previously been studied for systems of two immiscible liquids, one of which replaces the air in our setup[1,2]. The evolution we see here is very similar to this previously discovered cascade, but progresses at much higher speeds due to the negligible inertia of the surrounding medium. Furthermore, the satellite drops observed in the two-liquid case are rarely observed here. These are pinched off in the neck region and occur here only occasionally for the largest drops.

We have observed this phenomenon for liquids having widely different properties (water, alcohol and to lesser extent for mercury) and it should thereby be quite ubiquitous, but may have escaped much notice due to its rapidity, which makes the process barely perceptible to the naked eye. The six steps expire in about a quarter of a second. This cascade could be of considerable interest to the natural generation of mist and rain drops[3], bouncing of droplets[4,5], mixing at an interface[6] and the generation of vorticity[7–12]. Video images of natural rain do indeed show partial coalescences of the drop which is pinched off at the top of the Worthington jet.

The remarkable similarity of each step in the cascade is demonstrated in Fig. 1(b), which shows the pinch-off shapes for the same drop at the subsequent stage. The large number of steps also suggests a dynamic similarity. Such similarity arises from the assumption that the coalescence is governed solely by the surface tension and the inertia of the liquid. We thereby neglect viscous friction as well as gravity, which serves the sole purpose of bringing each generation of drops to the flat surface, as the daughter drops tend to bounce, with the smallest drops bouncing highest. The physical parameters of importance are therefore the strength of the surface tension σ, the size of the drop D and its density ρ. These quantities form a time-scale associated with the surface-tension driven distortions of the drop[8,9], i.e.

$$\tau_\sigma = \sqrt{\rho D^3/\sigma} \tag{1}$$

The problem has an inherent geometric similarity at every step. However, for complete similarity the same fraction of the drop volume must coalesce in each step of the cascade, requiring a characteristic velocity. The excess capillary pressure inside the drop ($\Delta p = 4\sigma/D$) will accelerate some fraction of the drops' mass ρL^3 during the characteristic capillary time τ_σ, which gives a velocity $U_\sigma = \tau_\sigma \Delta p D^2/(\rho L^3)$. Using this velocity we can estimate the relative importance of inertia and surface tension by the Weber number $We = U_\sigma\sqrt{\rho D/\sigma}$. Substituting for U_σ and using eq. (1) we obtain $We = D^3/L^3$ which is a constant, by assumption. The similarity of the cascade therefore follows directly from the validity of eq. (1), which is demonstrated for alcohol drops in Fig. 2. The duration of τ_σ was determined by counting frames from the high-speed video camera[13] operated at frame-rates as high as 40500 f/s. These extreme frame-rates are required for resolving the coalescence process for the smallest drops, which are less than 200 μm in diameter.

The horizontal drop diameters were measured from the video frames and corrected for the distortions due to gravity. This was done by measuring the volumes of a few different-size drops from images taken as they are pinched off from a nozzle. The horizontal diameters of the resting drops were then measured and a correction factor constructed to convert the horizontal diameter to the corresponding diameter of the same-volume sphere. This correction depends on drop size, but is only significant for the larger drops.

It remains an intriguing possibility[2] that further steps exist in such cascades, especially if those steps would generate drops too small for the current optical setup. This is however unlikely due to the damping effects of viscosity, the strength of which is characterized by the Reynolds number (UD/ν), where ν is the kinematic viscosity of the liquid. Its value can be estimated using the characteristic velocity as $U = D/\tau_\sigma$. Therefore, $Re \propto \sqrt{D}$ and for very small drops viscosity will inevitably start slowing down the process destroying the similarity. However, for the smallest drops observed here $Re \approx 100$ justifying the neglecting of viscous effects. Were one to arbitrarily select a Re of 20 as the cut-off where viscous forces become dominant, this would correspond to an alcohol drop of 8 μm in diameter.

It is worth noting that at high viscosities the pinch-off of a drop from a nozzle posesses a cascade of instabilities which have been investigated and successfully modeled[13,14].

High-speed video clips showing the present coalescence cascade are available at *http://www.tam.uiuc.edu/Faculty/Thoroddsen/Cascade.html*

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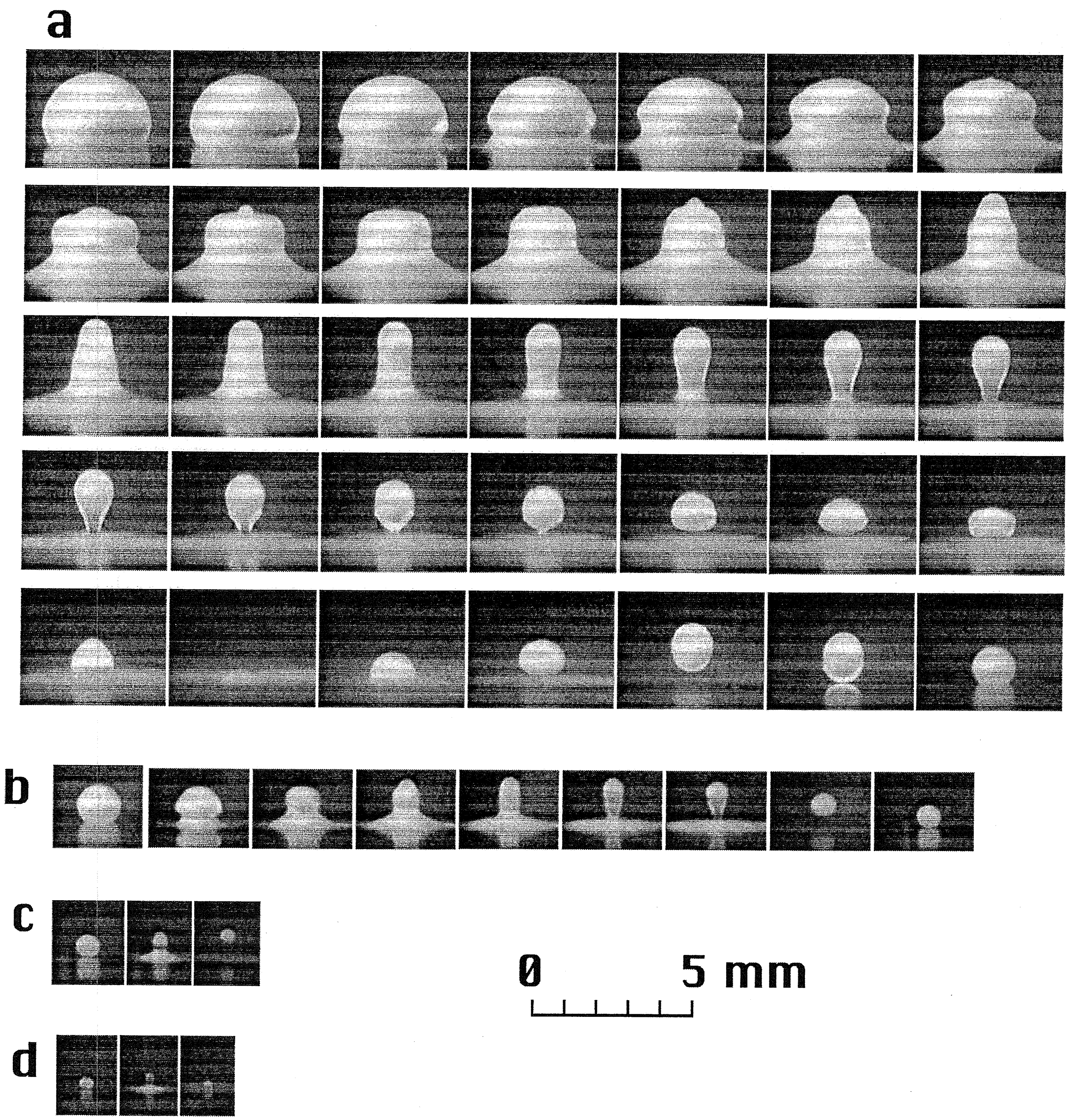

Figure 1: The coalescence cascade for a water drop. The drop contains a minute amount of Fluorescein for clearer imaging. Fluorescein was not used in obtaining the data in Fig. 2. **a**, The deformation and pinch-off of a daughter drop. The camera is operated at 2250 f/s. In the first four rows every frame is shown, whereas many frames have been left out between subsequent frames. **b**, The second step in the cascade, which begins about 70 ms after the end of the first sequence above. **c**, **d**, frames during the third and fourth steps.

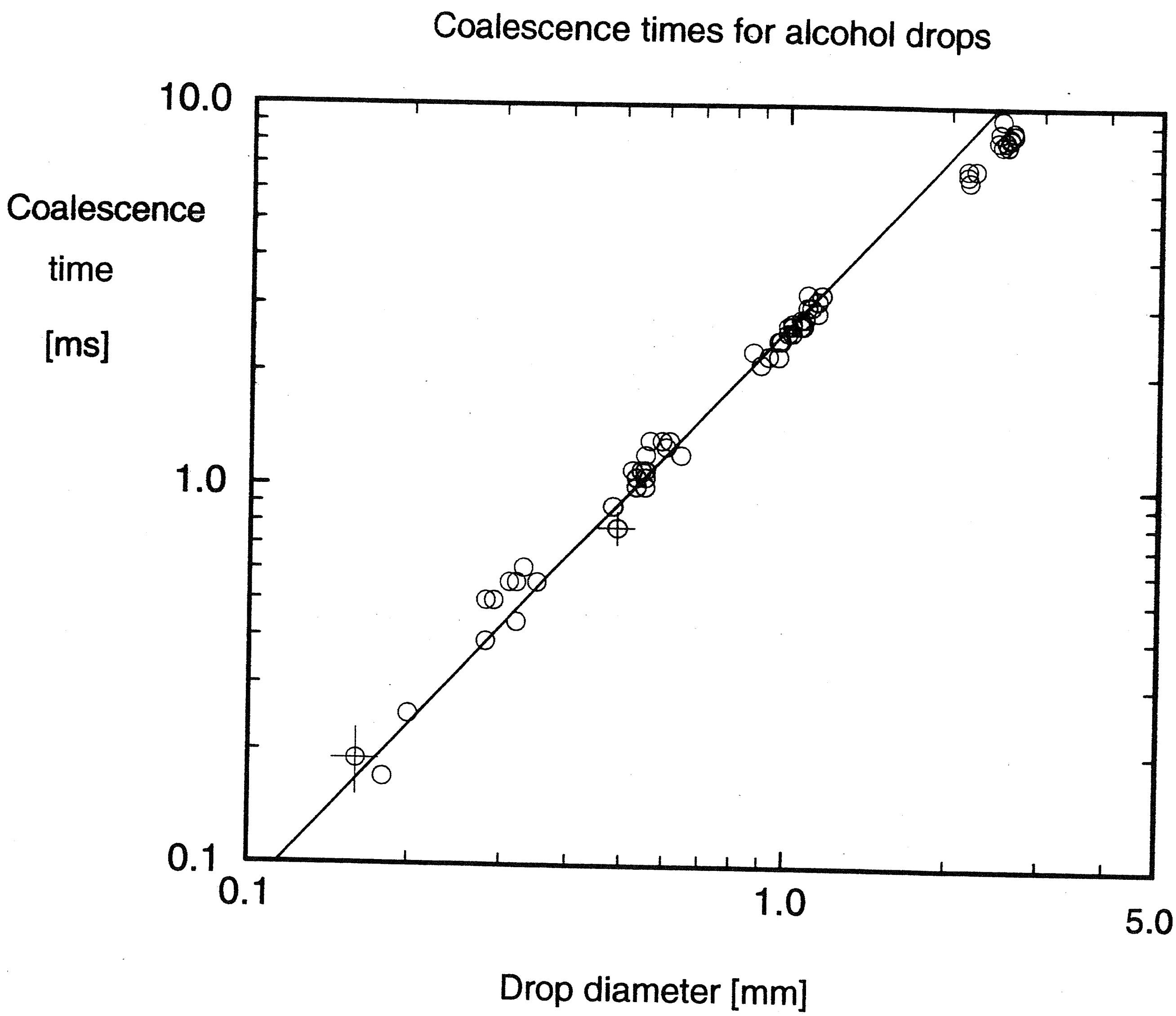

Figure 2: Log-log plot of the coalescence times for each step of the cascade, for different diameter drops of ethyl alcohol. τ_σ is estimated from first liquid contact to the pinch-off of the satellite drop. The initial diameters were determined from the video images, correcting for distortions due to gravity. The line has a slope of 3/2 confirming equation (1).

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