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Universal rim thickness in unsteady sheet fragmentation

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Unsteady fragmentation of a fluid bulk into droplets is important for epidemiology as it governs the transport of pathogens from sneezes and coughs, or from contaminated crops in agriculture. It is also ubiquitous in industrial processes such as paint, coating, and combustion. Unsteady fragmentation is distinct from steady fragmentation on which most theoretical efforts focused thus far. We address this gap by studying a canonical unsteady fragmentation process: the breakup from a drop impact on a finite surface where the drop fluid is transferred to a free expanding sheet of time-varying properties and bounded by a rim of time-varying thickness. The continuous rim destabilization selects the final spray droplets, yet this process remains poorly understood. We combine theory with advanced image-analysis to study the unsteady rim destabilization. We show that, at all times, the rim thickness is governed by a local instantaneous Bond number equal to unity, defined with the *instantaneous, local*, unsteady rim acceleration. This criterion is found to be robust and universal for a family of unsteady inviscid fluid sheet fragmentation phenomena, from impacts of drops on various surface geometries to impacts on films. We discuss under which viscous and viscoelastic conditions the criterion continues to govern the unsteady rim thickness.

Unsteady fluid fragmentation is ubiquitous and important for a wide range of industrial processes [1, 2], health applications [3–6] or pathogen dispersal in agriculture [7]. Fluid fragmentation generates numerous droplets that can travel over large distances and disperse their biological and chemical payloads. Prior work focused on characterizing the droplet size distribution as a function of the breakup geometry and fluid properties [2, 8, 9]. Theoretical insights focused on steady fragmentation, such as Savart sheets [9–11]. However, an important class of fragmentation processes are in fact *unsteady*: they continuously shed droplets with time-varying properties.

Unsteady sheet fragmentation occurs for crown splash upon drop impact on a thin liquid film [12–15] or deep pool [16, 17], or crescent-moon splash upon drop-drop interactions on surfaces [7, 18, 19]. These situations generate an expanding sheet of time-varying velocity and thickness profiles, bounded by a rim of time-varying thickness which destabilizes into droplets. The rim is the critical link between the sheet and droplets. **While linear instabilities of rims were extensively discussed [14, 20, 21],** time-resolved observations and theoretical insights on unsteady rim destabilization are lacking. Here, we present the first systematic demonstrations on the role of unsteadiness in rim destabilization.

Prior work on the canonical unsteady fragmentation process from drop impact on a surface of comparable size (Fig. 2a) [21–27], assumed that most droplets are shed after full radial sheet expansion. Our recent work, however, shows that the droplet size and speed distributions are determined during the sheet expansion [28], in which the *rim destabilization* plays a key role. Unsteadiness introduces a more subtle dynamics than the classic debated dichotomy between Rayleigh-Plateau [29–31] and Rayleigh-Taylor instabilities. [21, 32, 33]. A prior

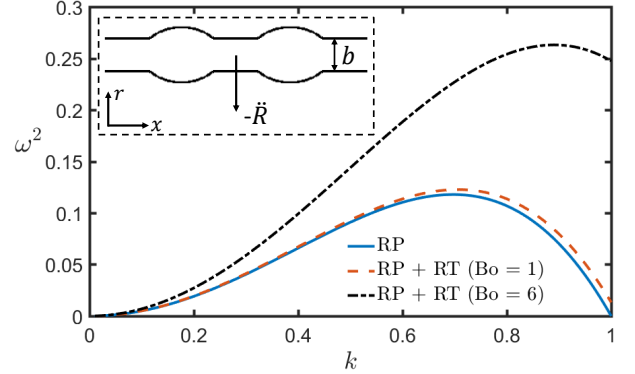


FIG. 1. Dimensionless dispersion relation of the coupled Rayleigh-Plateau (RP) and Rayleigh-Taylor (RT) instabilities for different Bond number Bo compared to that of the Rayleigh-Plateau instability. The inset shows a fluid cylinder of diameter b subject to an acceleration $-\ddot{R}$.

study [34] considered the interplay between the coupled Rayleigh-Plateau and Rayleigh-Taylor instability using linear stability analysis. Here we consider them jointly acting on an inviscid cylindrical liquid jet subject to an acceleration as shown in Fig. 1(inset). The dispersion relation of the coupled instability can be derived as [see Supplementary Information]:

$$\omega^2 = \frac{1}{2} \left(-\chi(k) + \sqrt{\chi(k)^2 - 4\psi(k)} \right),$$

$$\text{with } \chi(k) = \frac{kI_1(k)}{I_0(k)}(k^2 - 1) + \frac{kI_2(k)}{I_1(k)}k^2 \quad (1)$$

$$\text{and } \psi(k) = \frac{k^2 I_2(k)}{2I_0(k)} \left[2(k^2 - 1)k^2 - (Bo/4)^2 \right],$$

where $I_n(k)$ is the first kind modified Bessel function

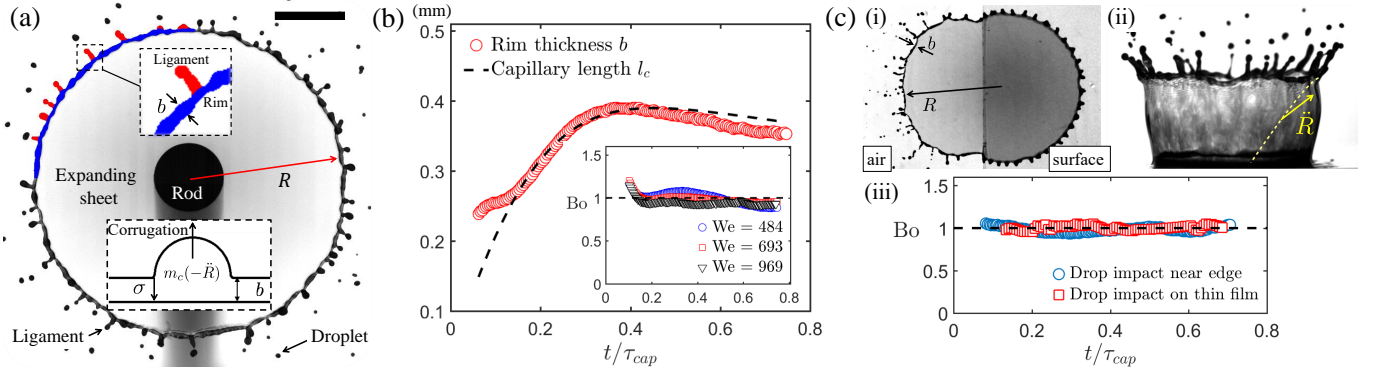


FIG. 2. (a) Unsteady sheet fragmentation upon drop impact on a target of comparable size which ensures an horizontal expanding sheet [22]. Scale bar is 6 mm. The upper inset shows the rim-ligament separation by our image processing algorithm. The lower shows the schematic diagram of a local corrugation. (b) Time evolution of the sheet rim thickness compared to the local instantaneous capillary length l_c based on the rim acceleration $-\ddot{R}$ for $We = 693$. Time is non-dimensionalized by the global capillary time $\tau_{cap} = \sqrt{\rho d_0^3 / 8\sigma}$ of sheet expansion, where d_0 is the diameter of the impacting drop. The inset shows the local, instantaneous Bond number of the rim $Bo = 1$, robust for three different Weber numbers. (c) **Sheet fragmentation from (i) impact near a surface edge, causing the sheet's left part to expand in the air; and (ii) drop impact on a thin liquid film with a crown rising upward.** (iii) The criterion of $Bo = 1$ holds for the rim in the air in case (i) and for the crown rim in case (ii).

of order n , k is the wavenumber non-dimensionalized by the rim radius $b/2$, and ω is the growth rate non-dimensionalized by the rim capillary timescale $\tau_c = \sqrt{\rho b^3 / 8\sigma}$, where ρ and σ are the density and surface tension of the fluid, respectively. $Bo = \rho(-\ddot{R})b^2/\sigma$ is the *instantaneous* and *local* rim Bond number based on the *instantaneous* rim thickness b and acceleration \ddot{R} . When $Bo = 0$, (1) simplifies to $\omega^2 = \frac{k I_1(k)}{I_0(k)}(k^2 - 1)$, the inviscid Rayleigh-Plateau instability dispersion relation. This remains true at first order for $Bo \sim O(1)$ (Fig. 1). Only for $Bo \gg 1$ does the dispersion relation deviate significantly from that of Rayleigh-Plateau, yet remaining distinct from that of the Rayleigh-Taylor instability of a planar sheet $\omega^2 = k((Bo/4)^2 - k^2)$.

The unsteadiness of the acceleration $-\ddot{R}$ aggravates the complexity of the rim destabilization. To gain key physical insights on its role, we developed advanced image-processing algorithms that capture the contour of the rim-ligament system, and separate the rim from the ligaments (Fig. 2a) to measure precisely the time-varying rim thickness b . Figure 2b shows that the rim thickness b matches very well with the time-varying capillary length $l_c = \sqrt{\sigma/\rho(-\ddot{R})}$ based on the *instantaneous* sheet acceleration $-\ddot{R}$, i.e., $b = l_c$. This leads to an important criterion: the *instantaneous, local* Bond number of the rim $Bo = \rho b^2(-\ddot{R})/\sigma = 1$ holds at all times. Moreover, such criterion is independent of the impact Weber number $We = \rho u_0^2 d_0 / \sigma$, where u_0 is the impacting velocity of the drop and d_0 its diameter (Fig. 2b-inset).

To rationalize the $Bo = 1$ criterion we first examine the force balance on a growing corrugation on the rim (Fig. 2a-lower inset). Distinct from a free liquid jet, the rim of an unsteady expanding sheet does not fragment

into droplets directly. Corrugations grow along the rim to form ligaments while fluid continuously enters the rim from the sheet at a time-varying rate. As a small corrugation grows to form a bulge of size proportional to the rim thickness b , in the non-inertial reference frame of the rim, the deceleration exerts a fictitious force on the bulge, pulling it away from the rim. Simultaneously, surface tension pulls the bulge toward the rim (Fig. 2a-inset). The resulting force balance on the bulge is

$$m_b(-\ddot{R}) \sim \sigma b \quad \Rightarrow \quad b \sim \sqrt{\frac{\sigma}{\rho(-\ddot{R})}}, \quad (2)$$

where $m_b \sim \rho b^3$ is the mass of the bulge. Thus, if the rim thickness is much larger than the local instantaneous capillary length, a bulge is pulled away from the rim.

We can compute the prefactor of the scaling law (2) combining the linear instability analysis discussed earlier with the local momentum conservation of the growing bulge. When $Bo \sim O(1)$ the modified dispersion relation (1) is close to that of the inviscid Rayleigh-Plateau instability (Fig. 1). The associated fastest growing wavelength is $\lambda_{max} = 2\pi/k_{max} \simeq 4.5b$. Considering a spherical bulge of volume equal to that contained within one wavelength of the fluid column with diameter $d_b = (27/4)^{1/3}b \simeq 1.9b$, and mass $m_b = \rho\pi d_b^3/6$, surface tension pulls on such bulge with a force $F_\sigma = \sigma\pi d_b$. The dimensionless growth rate of the fastest growing mode of the inviscid Rayleigh-Plateau instability is $\omega_{max} = 0.343$ (Fig. 1). Thus, the characteristic growth timescale of a corrugation into a bulge is $\tau_g = \tau_c/\omega_{max}$. Using the bulge radius $d_b/2$ as a characteristic lengthscale, the resulting growth speed is $v_b = d_b/2\tau_g$.

Momentum conservation on the bulge reads

$$m_b(-\ddot{R}) + \rho A_b v_b^2 = \sigma\pi d_b, \quad (3)$$

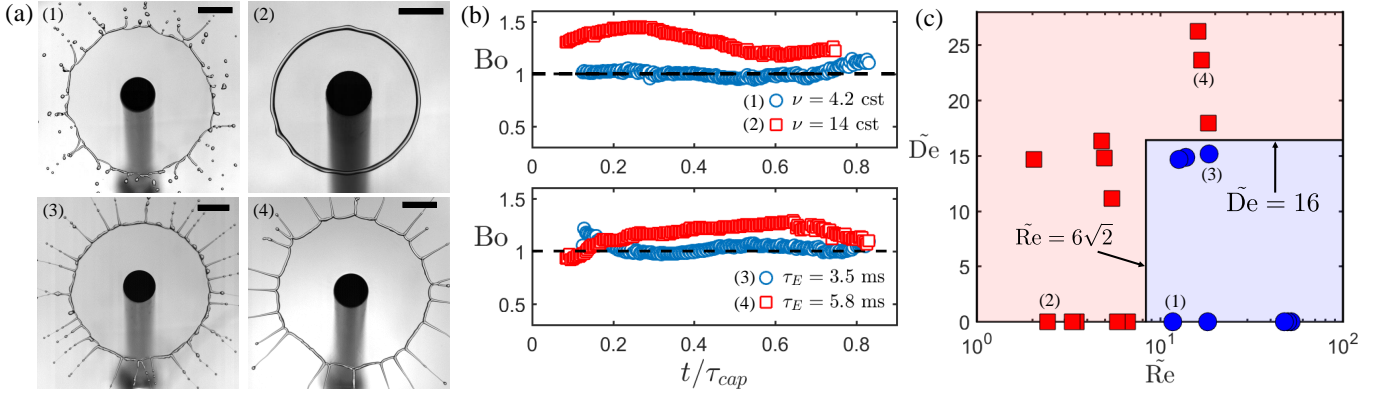


FIG. 3. (a) Sheet expansion upon drop impact on pole for fluids of different viscosity and elasticity. Scale bars are 6 mm. (b) Time evolution of the local instantaneous rim Bond number for the fluids shown in (a). (c) Regime diagram in terms of the local Reynolds number \tilde{Re} and Deborah number \tilde{De} defined in (5). Circles represent the experimental conditions for which $Bo = 1$ holds, and square the conditions for which it fails, for the rim of unsteady expanding sheet upon drop impact on a small surface. The criterion to determine if the $Bo = 1$ holds is that $0.8 < \langle Bo \rangle < 1.2$.

where $A_b = \pi d_b^2/4$ is the bulge cross-section area, giving

$$b \simeq \sqrt{\frac{\sigma}{\rho(-\ddot{R})}} \Rightarrow Bo = \frac{\rho(-\ddot{R})b^2}{\sigma} \simeq 1 \quad (4)$$

This result matches our data very well (Fig. 2b). Besides sheet fragmentation upon drop impact on a surface of size comparable to the drop, we also show that the criterion $Bo = 1$ applies to other fragmentation processes including **asymmetric sheet expansion in the air from a drop impact near a surface's edge** (Fig. 2c-i) and **for drop impacts on thin films leading to a crown** (Fig. 2c-ii). Despite changes in geometry and two-to-three dimensional sheet expansions, the unsteady local criterion $Bo = 1$ regulating the rim thickness holds. This means that the local unsteady rim thickness retains the value of the *local and instantaneous* capillary length throughout the unsteady sheet expansion. This remains true as long as sufficient fluid enters the rim **and viscous stresses are negligible** as discussed hereafter.

The criterion of instantaneous local $Bo = 1$ governing the unsteady rim thickness also applies to the fragmentation of fluids with a range of viscous and elastic properties summarized in Table 1 (Supplementary Information). Figure 3b shows the time evolution of the local instantaneous Bond number of the sheet rim for the same Weber of impact but different ν and τ_E . The $Bo = 1$ criterion continues to hold within critical regions of viscous and elastic effects (Fig. 3b,c). To better understand the effects of viscosity and elasticity, we examine the local rim Reynolds $\tilde{Re} = v_b b / \nu$ **in the non-inertial rim frame** (competition of inertial to viscous effects) and Deborah numbers $\tilde{De} = \tau_E / \tau_g$ (competition of elastic to capillary effects). Using the characteristic velocity $v_b \simeq \sqrt{\sigma / \rho b}$ and time $\tau_g \simeq \sqrt{\rho b^3 / \sigma}$ of rim destabilization, $\tilde{Re} = \sqrt{\sigma b / \rho \nu^2}$ and $\tilde{De} = \tau_E / \sqrt{\rho b^3 / \sigma}$.

Prior studies of drop impacts gave a maximum radius of the expanding sheet $R_{max} \sim \sqrt{We} d_0$. Thus, the sheet deceleration scales as $(-\ddot{R}) \sim R_{max} / \tau_{cap}^2 \sim u_0 / \tau_{cap}$, where $\tau_{cap} = \sqrt{\rho d_0^3 / 8\sigma}$ is the global capillary timescale characteristic of the sheet expansion. Experiments show that the variation in sheet deceleration during sheet expansion remains small compared to its mean value $\langle -\ddot{R} \rangle \simeq 2u_0 / \tau_{cap}$ (Fig. 2 in the Supplementary Information). Substituting this expression into (4) gives $b \simeq 0.2We^{-1/4}d_0$. Thus, the local Reynolds and Deborah numbers become

$$\tilde{Re} \simeq 0.2Oh^{-5/4}Re^{-1/4} \quad \text{and} \quad \tilde{De} \simeq 4DeWe^{3/8}, \quad (5)$$

where the Reynolds $Re = u_0 d_0 / \nu$, Ohnesorge $Oh = \sqrt{\rho \nu^2 / (\sigma d_0)}$, and Deborah numbers $De = \tau_E / \tau_{cap}$ are based on impact conditions. A regime map in terms of \tilde{Re} and \tilde{De} defined in (5) is shown in Fig. 3(c): **circles show experiments for which $Bo = 1$ holds and squares those for which it fails**. The region within which $Bo = 1$ holds is bounded by $\tilde{Re} \gtrsim 8$ and $\tilde{De} \lesssim 16$.

The dispersion relation $\omega = \omega(k)$ of the Rayleigh-Plateau instability used for the derivation of $Bo = 1$ only applies for inviscid fluids. The full relation accounting for both inertia and viscosity is implicit [35] but can be simplified asymptotically for small viscosity to yield the fastest-growing wavenumber $k_{max}^2 = 1 / (2 + 6\sqrt{2}/\tilde{Re})$ [36]. This shows that the inviscid dispersion relation no longer applies for $\tilde{Re} < 6\sqrt{2}$. **Thus, the $Bo = 1$ criterion should also fail when $\tilde{Re} < 6\sqrt{2} \approx 8.5$, which matches our experimental data very well** (Fig. 3c).

Prior studies on the Rayleigh-Plateau instability [37–41] showed that the breakup of a column of fluid with high elasticity is slower than that of a Newtonian fluid. This is consistent with our experimental data, which shows $Bo > 1$ for highly-elastic fluids, indicating that more fluid is accumulated in the rim due to less desta-

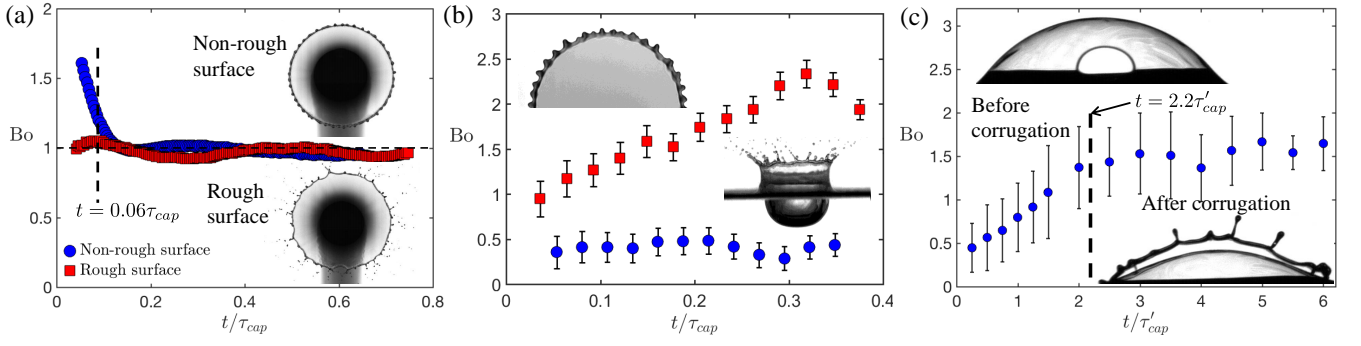


FIG. 4. (a) Time evolution of the local Bond number of the rim upon drop impact on a small smooth surface, compared with that upon impact on a rough surface for $We = 693$. The insets show the sheet at time $t = 0.06\tau_{cap}$, when the fragmentation is occurring on the rough surface while it has not yet started on the smooth one. (b) Time evolution of the local Bond number of the rim from drop impact on a deep pool for $We = 824$ (circles) and from impact on a solid surface for $We = 543$ (squares), showing that $Bo = 1$ fails in these two cases. (c) Time evolution of the local Bond number of the rim from bubble bursting. The capillary time in this case is $\tau'_{cap} = \sqrt{\rho(rh)^{3/2}/\sigma}$ with $r = 5.6$ mm and $h = 7.4$ μ m the bubble cap radius and thickness, respectively. The Bond number is computed based on the centripetal acceleration measured at the early stage of the hole retraction on the bubble cap.

bilization. However, the quantification of the nonlinear effects of fluid elasticity on rim destabilization and its modeling remain unclear. The experimental critical value of \tilde{De} above which $Bo = 1$ breaks down is $\tilde{De} \approx 16$ (Fig. 3c). The theoretical explanation for this value is the subject of future research.

We showed that the rim destabilization during unsteady sheet fragmentation induces a self-adjustment of the instantaneous rim thickness b : it remains equal to the local instantaneous unsteady capillary length. Namely, the rim thickness is governed by a *local* and *instantaneous* Bond number $Bo = \rho b^2(-\ddot{R})/\sigma = 1$. Such criterion is robust to a range of fragmentation geometries and to changes in fluid properties, including viscosity and elasticity. However, prerequisites need to be met to ensure that the $Bo = 1$ criterion shapes the rim thickness. First, the criterion only holds if sufficient fluid is contained in the rim. Second, initial perturbations need to be sufficient to trigger the unsteady instability ensuring growth of corrugations. This is illustrated in Fig. 4(a), showing the time evolution of the local rim Bo for drop impacts on smooth and rough surfaces. The former incurs less initial perturbations than the later, resulting in a longer time required for initial corrugations to grow into ligaments and, therefore, to reach $Bo = 1$. Third, the $Bo = 1$ criterion requires a sufficiently large acceleration and fluid influx into the rim. For instance, in the stationary case of a Savart sheet [42–44] the acceleration is null. Based on (4), the rim thickness would be infinite, clearly unrealistic. In this particular case the rim still destabilizes but its thickness is determined by the local geometry of cusps that develop around the rim [45].

For another unsteady sheet fragmentation, drop impact on a deep pool leading to a crown splash, we find that the local Bond number of the rim remains smaller

than 1 (Fig. 4b). This is due to the important dissipation of impact kinetic energy via cavity formation below the surface, which decreases the fluid influx into the rim below what would be required to satisfy $Bo = 1$. The physical implication of $Bo < 1$ is that, contrary to impacts on thin films or surfaces, corrugations cannot grow. When observed however, they appear to form at the very early stage of impact and are believed to originate from a different mechanism [31, 46–49]. The complexity of the early stage of impact makes their origin difficult to elucidate. However, with the instantaneous local $Bo < 1$, no new ligaments emerge from the rim during its expansion. This is in contrast to the continuous emergence of new ligaments from unsteady rims from drop impacts on surfaces and thin films with $Bo = 1$ (Fig. 2). For drop impact on large solid surfaces, viscous stresses stabilize the rim: it accumulates fluid that cannot be shed, thus, the local Bo is systematically larger than 1 (Fig. 2).

Beside unsteady expanding sheet fragmentation, fragmentation processes resulting from sheet rupture are also of interest. They arise, for example, from bursting bubbles [3] (Fig. 4c). The sheet retraction on a bubble cap is governed by the constant Taylor-Culick retraction speed [50, 51], yet the curvature of the bubble cap induces a centripetal acceleration normal to the rim direction of motion. The rim thickness is first *primarily* driven by the scavenging of fluid throughout the hole opening, independent of the acceleration, leading to $Bo < 1$. Here, Bo is defined based on the constant initial centripetal acceleration. As fluid accumulates into the rim, eventually, the local instantaneous Bo reaches 1, coinciding with the growth of corrugations in the direction of the centripetal acceleration. Beyond that point, caution in interpretation of the later stage is required as the bubble cap sheet curves with a kink forming in the direction

of the centripetal acceleration, in addition to no longer being a stretched film as the rest of the bubble collapses.

In the context of fluid fragmentation, our study represents a significant advance. Unsteady sheet fragmentation is ubiquitous in industrial and natural fluid processes, used for drug delivery [52], and important for transfer of biological and chemical compounds and pathogen spread. Prior studies neglected the role of unsteadiness on rim destabilization, in large part due to the lack of measurements enabling to discriminate between hypotheses on the dominant instabilities taking place, but also due to the lack of a theoretical framework as tractable as that of steady classical hydrodynamic instabilities. We showed that a robust criterion of instantaneous local $Bo = 1$ governs the unsteady rim thickness. This criterion enables tremendous simplification of the modeling, and therefore the mechanistic understanding, of a large class of unsteady inviscid fragmentation processes. Moreover, we also showed for which fluid viscous and viscoelastic conditions the criterion continues to hold, thus allowing its application to a broader class of contaminated and non-Newtonian fluids important in biological processes such as violent exhalations or crop pathogen transport involved in disease transmission [4–7].

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