Supplementary material on

"Electric-field mediated morpho-dynamic evolution in drop-drop coalescence phenomena in the inertio-capillary regime"

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S1. Effect of capillary wave propagation over father drop on pinch-off

As discussed in the main text, after attaining the maximum height, the protrusion formed at the apex of the mother drop undergoes vertical and horizontal collapse. If the horizontal collapse prevails (or the vertical collapse is delayed), the pinch-off occurs. Hence, it appears that the capillary wave propagation on the father drop plays an insignificant role in pinch-off, as the same primarily occurs due to morpho-dynamic evolution of the mother drop. This typically features for drops having large diameter ratio as for these cases, the capillary waves travel by a small distance so as to bring in small deformation to the father drop, before the vertical collapse sets on the mother drop (refer to figure S1(a) given below). Hence, the pressure change inside the father drop becomes negligible and, accordingly, the pressure-driven fluid drainage from the mother to father drop that governs the horizontal neck collapse remains almost unaffected. However, for farther drops of size close to the size of the mother drops (i.e., for small diameter ratio), the capillary wave propagates to a significantly larger extent in the father drop, causing major distortion in its shape, which can influence the pinch-off mechanisms discernibly (refer to figure S1(b)).

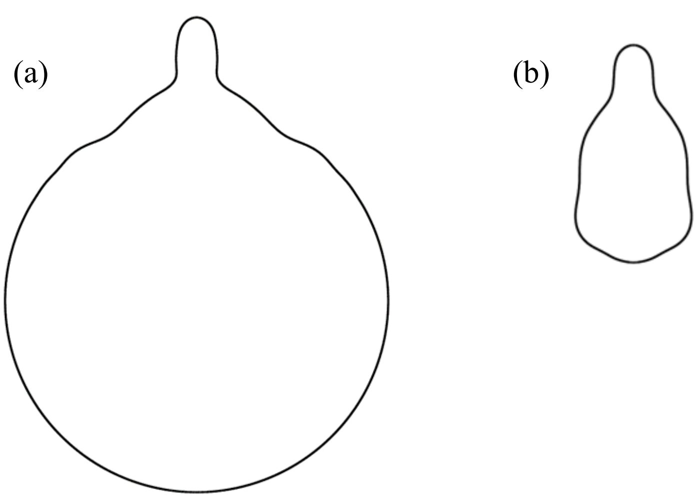

Figure S1. (a) Drop contour during the attainment of maximum height for (a) $\beta=5$ and (b) $\beta=1.6$.

Some insight into the effect of the wave propagation on the father drop on pinch-off can be obtained from the recent work of Deka *et al.*(2019). By interrupting the simulation after the mother drop stretched up to a maximum height, they set the velocity to zero in the bottom half of the drop, and could realize pinch-off after the same. This indicates that the wave propagation on the father drop is likely to hinder the pinch-off. However, considering the interplay of all the forces in an electrohydrodynamically modulated system as addressed herein, the effect of capillary-driven deformation of the father drop does not appear to be consequential as compared to the effect the viscous forces and the electrically-modulated forces at the contact region that are tunable to a large extent by varying the diameter ratio.

S2. Difference between the coalescence in drop-drop system and drop-planar surface

We first highlight the difference between the coalescence in drop-planar interface and drop-drop coalescence, in the absence of electric field, as exemplified in figure S2. To this end, we compare our numerical results (colored outlines) with the reported experimental results of Blanchette & Bigioni (2006). In their experiments, ethanol drop of diameter of 1.06 mm was allowed to fall through air onto the reservoir of an identical liquid in the absence of electric field. Executing simulations for diameter ratio β=1.6 and 5, we observe that for β=1.6, the numerical results agree with the experimental results to some extent at early times, but progressively tend to deviate. At 3.57 ms, the onset of pinch-off can be observed in the experiments but not in the simulations. Increasing the diameter ratio to β=5, however, results in significantly closer agreement between the experiments and the simulations. This may be attributed to the fact that increase in diameter ratio increases the surface flatness of the father drop near the contact region. Thus, for large diameter ratio (β>>1), a drop-drop system can approximately behave as a drop-reservoir system.

Next, we illustrate the role of electric field in altering the topological evolution of the drop over time (refer to figure S3). For this purpose, we compare our results with the reported experimental results of Mousavichoubeh *et al.* (2011). Their experiments were conducted using

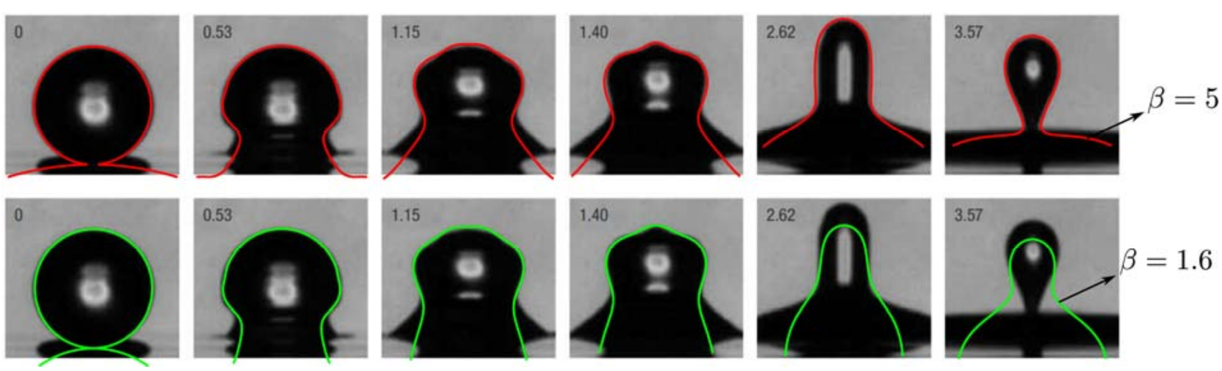

Figure. S2. The coalescence of drop-drop system and drop-interface system in the absence of electric field. Two drop-drop systems with β=1.6 and 5 are considered. Other parameters considered are Oh_i=0.011, Oh_e=0.0005, Bo=0.38 and A=0.997.

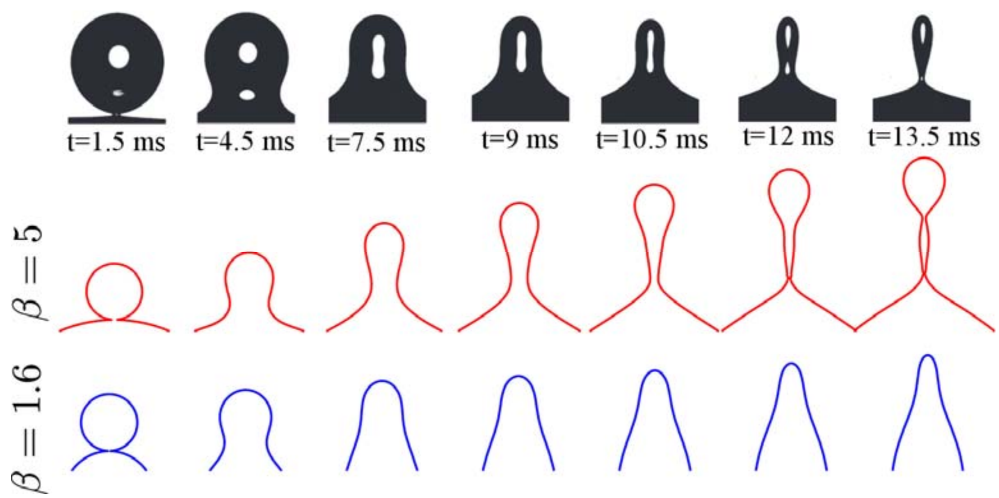

Figure. S3. The coalescence of drop-drop system and drop-interface system under electric field. Two drop-drop systems with β=1.6 and 5 are considered. Other parameters considered are R=100, S=16, Oh_i=0.006, Oh_e=0.3, Bo=0.04 and A=0.04.

water drop $\left(\sigma \sim 10^{-6} Sm^{-1}, \varepsilon \sim 80\varepsilon_0\right)$ and sunflower oil medium $\left(\sigma \sim 10^{-11} Sm^{-1}, \varepsilon \sim 4.9\varepsilon_0\right)$ placed above a water reservoir. The other parameters are: Oh_i=0.0064, Oh_e=0.3, Bo=0.04, A=0.04 and Ca_E =0.29. It can be seen that both for β=1.6 and 5, the results for drop-drop system significantly deviate from the corresponding ones obtained for the drop-reservoir system. For β=1.6, the drop elongates along with continued merging, and finally a highly elongated drop is formed. For β=5, although the shape evolution is qualitatively similar, the pinch-off results in a much larger daughter drop as compared to the daughter drop obtained in case drop-reservoir system. The reasons behind the deviation are as follows.

For a drop-reservoir system, the flat interface does not deform perceptibly. Hence, the electric field distribution around the drop evolves in repose to its own shape alteration only, unlike the deformable body pair formed by the drop-drop system. For a given electrical capillary number, the equivalent capillary number for the father drop is more, as discussed in the section 5.1 of the main text. For the capillary number considered herein, the father drop elongates vigorously in the direction of the electric field, distorting the electric field significantly. Moreover, the elongation of the father drop may stimulate fluid flow towards its ends (one end forming the contact region itself), which may largely inhibit the fluid drainage from the mother to the father drop. This brings in contrasting features of the drop-drop electrohydrodynamic system as compared to the drop-interface electrohydrodynamic system that hitherto remained unaddressed.

Reference

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