Supplementary Material: Inertial stretching separation in binary droplet collisions

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I. MORE ON THE EFFECT OF THE OFFSET ON THE MAXIMUM DEFORMATION LENGTH

To track the dynamics of the stretched system, we track the change in its normalised major length (\tilde{L}) over time. The normalised major length is defined as the longest line that can be bounded by the edge of the system of the coalesced droplets normalised by the original diameter of the droplets. Fig. S1 is an example that shows how \tilde{L} evolves over time for 8% HPMC (Oh = 0.214) at We = 83 for different offsets. As it can be seen, For B ≤ 0.38 the system stretches to its maximum (\tilde{L}_{max}) then retracts. The stretching separation occurs if $\tilde{L}_{max} \geq \tilde{L}_{max,c} = 3.35$, as in the collisions of B = 0.31, 0.33 and 0.38. However, in collisions with B significantly greater than B_c (here in this example B_c = 0.31), such as in B ≥ 0.41 , the system keeps stretching until separation without any retraction. Finally, Fig. S1 also shows how comparable \tilde{L}_{max} in the collisions of B ≤ 0.2 that implies the negligible effect of B at this range, seen in the region I in Fig. 4a in the main paper.

II. MORE ON THE DERIVATION OF THE C-SS TRANSITION MODEL

The dynamics of head-on binary droplet collisions have been studied by many authors [1–5]. Based on energy balance in the compression phase (between the initial instant of collision and the instant of maximum deformation, as shown in Fig. S2), Planchette *et al.* [2] demonstrated that there is a fixed ratio (α) of viscous loss ($E_{\mu,comp}$) to the initial kinetic energy ($E_{k,ini} = \pi \sigma D_o^2 We/24$), as for a given Oh, α is constant for a wide range of $E_{k,ini}$ (i.e. We) including the range covered in this work, as demonstrated in Fig. S2. However, the value of α depends on Oh, as shown in figure 7 in the main paper. At low Oh values (≤ 0.02), α generally increases with Oh from 0.50 to 0.65 ± 0.05 . For Ohnesorge numbers from 0.02 to 0.14, α remains constant at 0.65 ± 0.05 , therefore exhibiting *inertial behaviour* (*independent of viscosity*). For Oh > 0.14, the value of α is scattered but with a value generally above that of the preceding constant region. Extensive details and scaling arguments for why these three behaviors of α are seen can be found in Ref. [2].

Using the above knowledge of the viscous loss, the surface energy at the maximum deformation $(E_{\sigma,max})$ can be predicted via the energy balance at the compression phase $E_{\sigma,max} = (1 - \alpha)E_{k,ini} + E_{\sigma,ini}$, where $E_{\sigma,ini}$ is the initial surface energy of the droplets and is given by $E_{\sigma,ini} = 2\pi D_o^2 \sigma$. Using volume conservation, Willis and Orme [3] reported that the shape of the system at the maximum deformation can be well approximated by a pancake-shape of diameter $L_{max}|_{B=0}$ and thickness $\sim 4D_o^3/3L_{max}^2|_{B=0}$. Hence, its surface energy is given by

$$E_{\sigma,max} = \frac{\pi}{2} \sigma L_{max}^2 \Big|_{=0} \left(1 + \frac{8}{3} \left(\frac{D_o}{L_{max}} \Big|_{B=0} \right)^3 \right).$$

This equation is the same one used by Planchette *et al.* [2], and is used in this work to estimate α from the energy balance. Thereby, from the energy balance, $\widetilde{L}_{max}|_{B=0}$ can be estimated, as a function of the experimentally measured

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FIG. S1. The evolvment of the normalised major length of the coalesced droplets with time for collisions of 8% HPMC droplets (Oh = 0.214) at We = 83 at different offset. The time, here, is normalized by the droplet oscillation time $(T = (\pi/4)\sqrt{(\rho D_o^3)/\sigma})$. Here, filled symbols mean permanent coalescence whereas the open symbols mean stretching separation.



FIG. S2. A schematic defining the compression and the relaxation phase in head-on collision.



FIG. S3. The viscous loss in the compression phase as a function of the initial kinetic energy at head-on collision for three different Oh. Note that the intercept in all the fitted lines here is zero.



FIG. S4. Influence of the impact parameter, B, on the maximum length that the coalesced droplets attain.

 α and We, from

$$\widetilde{L}_{max}\big|_{B=0} = \sqrt{\frac{(1-\alpha)We + 48}{12\left(1 + \frac{8}{3\widetilde{L}_{max}^3}\Big|_{B=0}\right)}}.$$
(1)

III. EXAMPLE DATA OF WATER AND GLYCEROL

Fig. S4 shows that the far ends of our data water and 83% glycerol are consistence with the trends seen in figure 6a in the main paper.

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