Journal Name

ARTICLE TYPE

Cite this: DOI: 10.1039/xxxxxxxxxx

Marangoni-driven spreading of miscible liquids in the binary pendant drop geometry

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Supplementary material

0.1 Range of control parameters

	drop 1		drop 2		drops
	Surface tension (mN m^{-1})	Viscosity (mPa s)	Surface tension (mN m^{-1})	Viscosity (mPa s)	Initial diameter (mm)
Range	46.2 to 72.8	1 to 100	22.9 to 46.2	1 to 100	2.00 to 2.92
Error	5%	2%	5%	2%	0.1%

Table 1 Range of the material parameters. The properties of ink-containing liquids were measured as described below; literature values were taken for the other liquids[1–4].

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0.2 Surface tension measurements

Surface tension was measured in triplo, using a pendant drop system (Data Physics OCA 15EC). The surface tension as a function of the ethanol percentage is shown in figure 1; the dependency of the surface tension on the glycerol percentage is shown in figure 2, and the time-insensitivity of the surface tension is shown in figure 3.

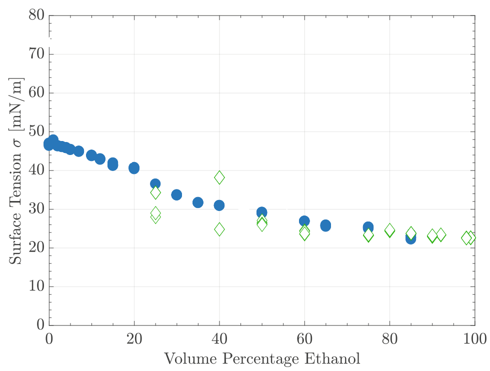

Fig. 1 (Color online) Surface tension of the water-ink-ethanol mixture as a function of the vol% of ethanol (●). 15 vol% ink (Brother LC-800) was dissolved. Literature values of water-ethanol mixtures without ink are from [2] (◇).

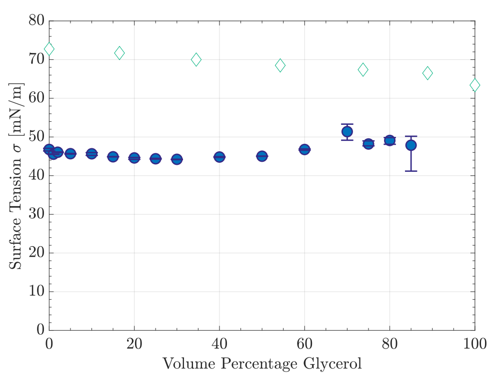

Fig. 2 (Color online) Surface tension of the water glycerol mixture, depending on the vol% of glycerol. For the values used in the paper (●) 15 vol% ink (Brother LC-800) was solved in the glycerol. Water-glycerol values are taken from Takamura *et al.* [4] (◇).

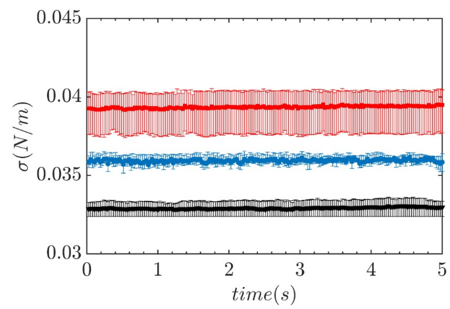

Fig. 3 (Color online) Surface tension of various mixtures as a function of time. 15% ink, 10% ethanol and 75% water (red), 100% ink (blue), 15% ink and 85% ethanol (black).

0.3 Viscosity measurements

Viscosity data was obtained using a rheometer (Anton Paar MCR 502), or from the literature, when indicated.

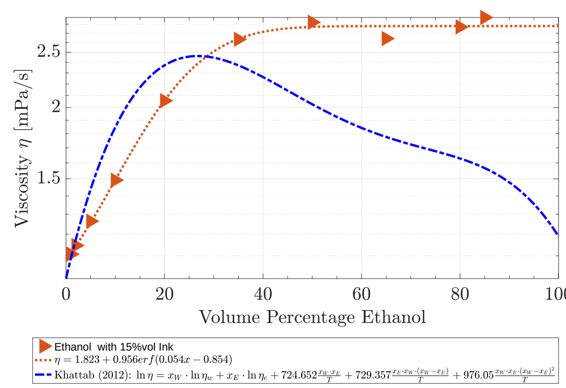

Fig. 4 (Color online) Viscosity of of the water ethanol mixture, depending on the vol% of ethanol. Measurements (▶) and data fit (- - -). 15 vol% ink (Brother LC-800) was solved in the ethanol. Water-ethanol mixtures from ref.[3] are shown by the blue line (-·-·).

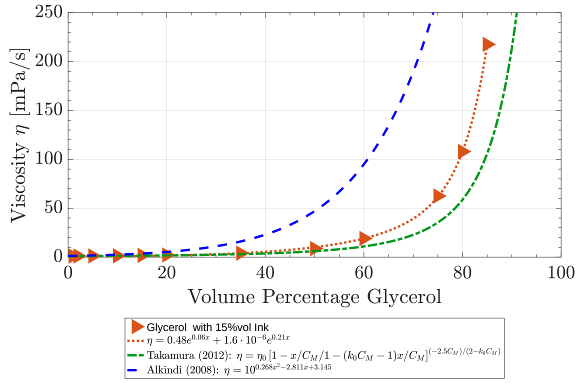

Fig. 5 (Color online) Viscosity of the water glycerol mixture as a function of the vol% of glycerol. Measurements (▶) and data fit (- - -). 15 vol% ink (Brother LC-800) was solved in the glycerol mixture. Previously determined values for water-glycerol mixtures are from[4] (-·-·) and[2] (- - -).

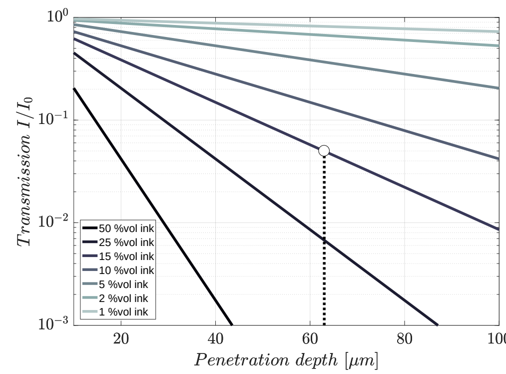

Fig. 6 (Color online) Optical transmittance of ink (Brother LC-800) in water solutions versus the penetration depth. 5% transmittance was chosen as the experimental threshold, corresponding to a penetration depth of 63μm shown by the symbol (○).

0.4 Optical penetration depth

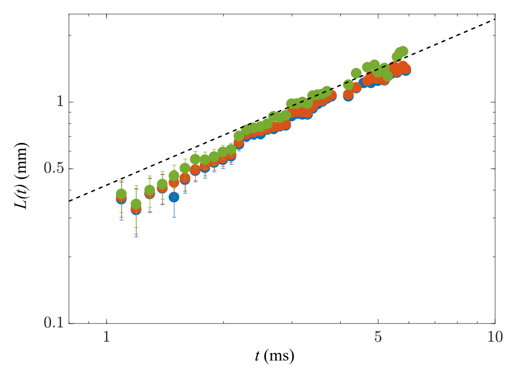

Fig. 7 (Color online) Effect of the threshold value on the measured spreading distance, $\eta_1 \approx \eta_2 \approx 1.5$ mPa s, $\Delta\sigma = 32$ mN m^{-1}. Optical penetration depths are 78 μm (●), 63 μm (●), and 51 μm (●). The dashed line (- - -) indicates $L(t) = 0.5\beta t^{0.77}$, $\beta = \Delta\sigma^{1/2}(\rho_1\eta_1)^{-1/4}$.

To determine the spreading rate, a threshold value for absorbed light has been chosen in the analysis. The transmission of light versus the optical penetration depth for the ink solutions are measured using a UV-VIS spectrometer (DR5000, Hach Lange), with pure air and a cuvette with water as references. The results are shown in figure 6. For 5 vol% ink a range of transmission has been chosen between $0.025 < I/I_0 < 0.1$, corresponding to optical penetration depths of 51µm to 78µm. The effect of the total transmittance threshold on the measurements of $L(t)$ is shown in figure 7. Increasing the transmittance leads to slightly lower penetration depth, therefore showing faster spreading. The effects in the ranges studied show the same exponents, $\alpha = 0.77$, and prefactors, 0.45β, for penetration depths of 63µm and 78µm. For the smaller penetration depth of 51µm, the prefactor increases to 0.51.

0.5 Boundary layer thickness

The transition from the deep-bath regime to the thin-film regime can be expected as soon as a backflow will limit the development of the boundary layer in drop 1. To first approximation, the viscous boundary layer cannot grow any further when $\delta_{BL} > D/4$, as shown in Figure 8. The Blasius boundary layer thickness is described by $\delta_{BL} = (\eta t/\rho)^{1/2}$, as shown in figure 9b. Only for experiments with a highly viscous inner drop ($\eta_1 > 40$ mPa s), the boundary layer reaches $D/4$ as shown in Figure 9b. Indeed, in that case we observe a flattening of the spreading curve (Figure 9a) that is consistent with $\alpha = 1/2$ as expected for thin-film spreading[5].

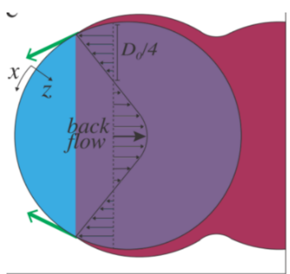

Fig. 8 (Color online) Optical transmittance of ink (Brother LC-800) in water solutions versus the penetration depth. 5% transmittance was chosen as the experimental threshold, corresponding to a penetration depth of 63μm shown by the symbol ($\circ$).

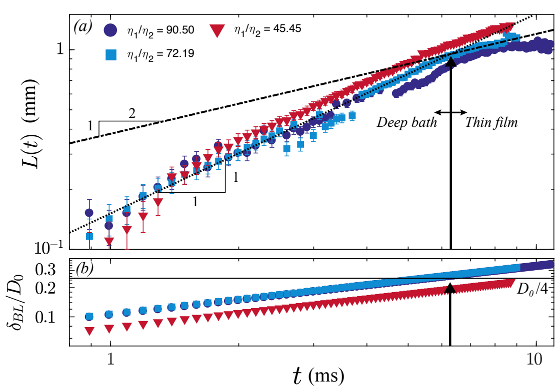

Fig. 9 (Color online) Temporal transition in spreading regimes; (a) Spreading versus time for data with large η_1/η_2. Initial spreading scales as $L(t) \sim t^{\alpha}$ with $\alpha = 1$. Later in time, a transition to $\alpha = 0.5$ is visible. Both slopes are given by the dashed-dotted (1/2) and dotted line (1); (b) Corresponding growth of relative boundary layer δ_{BL}/D_0. Transition to $\alpha = 0.5$ occurs around $\delta_{BL}/D_0 = D_0/4$, suggesting thin film-limited spreading when the boundary layer interacts with the toroidal vortices.

Notes and references

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