Electronic Supplementary Information (ESI)

Lateral migration of viscoelastic droplets in a viscoelastic confined flow: role of discrete phase viscoelasticity

Shamik Hazra¹, Sushanta K. Mitra² and Ashis Kumar Sen^{1,*}

¹Department of Mechanical Engineering, Indian Institute of Technology Madras, Chennai-600036, India. ²Waterloo Institute for Nanotechnology, University of Waterloo, Ontario N2L 3G1, Canada.

*Author to whom correspondence should be addressed. Email: ashis@iitm.ac.in

S.1 Properties of the fluids

Liquids	Density	Viscosity (mPa-s)	Viscosity ratio_w.r.t	IFT w.r.t PDMS	IFT w.r.t castor oil	Relaxation time(s)	Elasticity ratio w.r.t
	(gm/e///	(PDMS (k)	(mN/m)	(mN/m)		PDMS (ξ)
PVP 3%	1.0047	12.725	52.32	17.83-	17.20-	0.5[Go et al. ¹]	0.002
				20.02	18.48		
PVP 5%	1.0087	35.966	18.52			2.2×10^{-3}	0.45
						[Liu et al. 2],	
						1.6×10^{-3}	
						[Naillon et al. ³]	
PVP 6%	1.0107	57.154	12.0	16.03-	20	$\sim 9.4 \times 10^{-4}$	1.06
				17.31		[Yang et al. ⁴]	
PVP 9%	1.017	177.965	3.74			$\sim_{3.0\times} 10^{-3}$	0.33
						[Romeo et al. ⁵]	
PVP 10%	1.0187	257.695	2.58	20.3	17.80	6×10 ⁻³	0.16
						[Naillon et al. ³]	
PVP 13%	1.0239	654.67	1.01	16.05-			
				19.71			
PEG 15%	1.0293	57.655	12	21-25	16-20	~ 0	
PDMS	1.0051	666				$\sim 10^{-3}$	
Castor oil	0.960	650				~ 0	

Table S1: Measured properties of the fluids use in our experiments.

S.2 Rheometry data

Fig S1 summarizes rheometry results. Viscosities of different concentrations of PVP remain constant w.r.t strain rate (Fig S1a). The same holds true for PDMS base and PDMS 1.5:1 mixture (Fig S1b). Oscillatory shear test of PDMS 1.5:1 reveals very high value of storage modulus, which indicates strong elastic property of cross-linked PDMS (Fig S1c).



Fig. S1: (a) viscosity vs. strain rate plot of PVP 3%, 5%, 6%, 9% and 10%, (b) viscosity vs strain rate plot of PDMS base and 1.5:1 PDMS, (c) variation of storage modulus (\vec{G}) and loss modulus (\vec{G}) of PDMS 1.5:1 with frequency.

S.3 Empirical modeling and analytical scaling of F_{vd}

 F_{VD} originates from discrete phase viscoelasticity. Hence, it must be a strong function of relaxation time of the droplet phase, λ_D .

From experiment it is evident that F_{VD} is a function of viscosity ratio $(k = \frac{\mu_D}{\mu_M})$ as PDMS droplets are reversing the direction of migration for a range of k. Moreover, F_{VD} is a function of λ_M too as experiments revealed that reversal won't be observed in Newtonian continuous phase for the same viscosity ratio. F_{VD} has a complicated dependency on droplet diameter (D) which in turn is a function of continuous phase flowrate (strain rate, $\dot{\gamma}$). Fig S2 elaborates the effect of droplet size on lateral position in PVP 6%. For example, as we decreased the PVP 6 w/w % flowrate keeping PDMS flowrate constant, increasingly bigger PDMS droplets could be generated and their lateral equilibrium position shifted more towards the wall (Fig S2). Hence, for $\dot{\gamma}_1 > \dot{\gamma}_2 > \dot{\gamma}_3$ we get $r_1 < r_2 < r_3$ and $\Delta_1 > \Delta_2 > \Delta_3$, where, Δ_i is the film thickness separating PDMS droplet surface and the wall for corresponding droplet size r_i generated at $\dot{\gamma}_i$. From the above discussion F_{VD} takes the following functional form:

Instead of Δ_i we choose wall to droplet center distance, h to follow the convention in literature.



Fig. S2 (a-c) Decrease in film thickness with increase in PDMS droplet diameter in PVP 6%. (d) plot of film thickness vs. PDMS droplet diameter.

Using Buckingham's Pi theorem, we get five non dimensional numbers and Eq. (S1) can be expressed as

$$f\{\left(\mu_D/\mu_M\right), \left(D/h\right), \left(\lambda_D \dot{\gamma}\right), \left(F_{VD}/\mu_D D^2 \dot{\gamma}\right), \left(\lambda_D/\lambda_M\right)\} = 0$$
(S2)

To determine the interrelation among these five non-dimensional numbers we studied droplet dynamics in 300, 500 and 800 μm channel thereby changing the strain rate for same PVP flowrate. For each channel, flowrate was varied from 4-16 $\mu l/min$ with a step of 4 $\mu l/min$ and for each case same-sized droplets of both PDMS and castor oil were generated. For the same size, we propose that F_{VD} should be the difference of drag between PDMS and castor oil while they migrate laterally at different rates. Drag for flow past a viscous drop is given by ⁶

$$F_{Drag} = 3\pi\mu_{M}Dv \left(\frac{1 + \frac{2\mu_{M}}{3\mu_{D}}}{1 + \frac{\mu_{M}}{\mu_{D}}}\right)$$
(S3)

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The viscoelastic force due to discrete phase viscoelasticity, F_{VD} in Equation S2 can be calculated using Equation S3. Hence,

$$F_{VD}^{+} = F_{Drag,PDMS} F_{Drag,castor} = 3\pi\mu_{M}D(v_{pdms} v_{castor}) \left(\frac{1 + \frac{\mu_{M}}{3\mu_{D}}}{1 + \frac{\mu_{M}}{\mu_{D}}}\right) > 0$$
(S4)

$$v_{castor} = F_{drag,PDMS} F_{drag,castor} = 3\pi\mu_M D(0 - \frac{1 + \frac{2\mu_M}{3\mu_D}}{1 + \frac{\mu_M}{\mu_D}} < 0$$
(S5)

and for PVP 6 w/w

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for PVP 3 and 10 w/w %

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