Electronic Supplementary Material (ESI) for Lab on a Chip.

Electronic Supplementary Information

for

"Drop-of-Sample Rheometry of Biological Fluids by Noncontact Acoustic Tweezing Spectroscopy"

by

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Section S1. Theory for drop shape oscillation

Small amplitude axisymmetric deformations and shape oscillations of the viscoelastic liquid drop in microgravity were analyzed using the linear momentum and continuity equations as well as the Kelvin-Voigt constitutive equation[1] for drop viscoelasticity:

$$\tau = 2(G\gamma + \mu\dot{\gamma}),\ \ \dot{\gamma} = (\nabla\mathbf{v} + \nabla\mathbf{v}^{\mathrm{T}})/2,\ \ \gamma = (\nabla\mathbf{u} + \nabla\mathbf{u}^{\mathrm{T}})/2, \qquad \text{(S1)}$$

where τ is the deviatoric stress tensor, γ the strain tensor, $\dot{\gamma}$ the rate-of-strain tensor, $\mathbf{v}$ the velocity vector, $\mathbf{u}$ the displacement vector, ∇ the gradient operator, G the elastic modulus, and μ the shear viscosity. Assuming a negligible stress tensor in the medium outside the drop surface, the following kinematic and dynamic boundary conditions were applied at the drop surface, respectively:

$$\mathbf{v}_{\mathrm{s}} = \mathbf{v}|_{\mathrm{s}}, \qquad (p\mathbf{n} - \mathbf{n}\cdot\tau)_{\mathrm{s}} = \sigma(\nabla_{\mathrm{s}}\cdot\mathbf{n})\mathbf{n}, \qquad \nabla_{\mathrm{s}} \equiv \nabla - \mathbf{nn}\cdot\nabla. \qquad \text{(S2)}$$

Here $\mathbf{n}$ is the outward unit normal vector, $\mathbf{v}_{\mathrm{s}}$ the velocity vector at the surface, ∇_{s} the surface gradient operator, p the pressure, and σ the surface tension.

The analysis of drop oscillation follows our previous work[2] in which drop viscoelasticity was described by Jeffreys constitutive equation.[3] In this analysis, the surface profile of the drop r_{s} is expressed in polar coordinates (r, θ) as

$$r_{\mathrm{s}} = R[1 + \varepsilon C_n P_n(\cos\theta)e^{-\alpha_n t}], \qquad \alpha_n = \delta + i\omega, \qquad \text{(S3)}$$

where R is the drop radius, ε represents a small parameter that measures the amplitude of the drop deformation during axisymmetric shape oscillations, δ the decay factor, ω the frequency of shape oscillation, $P_n(\cos\theta)$ the Legendre polynomial, C_n an unknown coefficient, and n the mode of oscillation (n = 0 corresponds to radial oscillation without a change in shape, 1 pure translational motion of the drop, and 2 quadrupole shape oscillation). From Eq. (S3) it follows that p, $\mathbf{v}$, and τ are proportional to $e^{-\alpha_n t}$, with time-dependent coefficients of proportionality.[2] The displacement vector $\mathbf{u}$ can then be calculated from the velocity vector $\mathbf{v}$ by time integration of only $e^{-\alpha_n t}$. In this case, the Kelvin-Voigt model [Eq. (S1)] reduced to the Newtonian fluid model with effective viscosity:

$$\tau^{(n)} = 2\mu_{eff}\dot{\gamma}^{(n)}, \quad \mu_{eff} = \mu - G\alpha_n^{-1} \tag{S4}$$

The general solution for p, **v**, and τ looks like:

$$\begin{aligned}
p(r,\theta,t) &= \frac{2\sigma}{R} + \epsilon\rho\omega_L^2 R^2 \left[A_n\left(\frac{r}{R}\right)^n\right] P_n(\cos\theta)e^{-\alpha_n t}\,, \\
\boldsymbol{v}_r(r,\theta,t) &= \epsilon\omega_L R\left[nA_n\frac{\omega_L}{\alpha_n}\left(\frac{r}{R}\right)^{n-1} + \frac{B_n R j_n(kr)}{r}\right] P_n(cos\,\theta)e^{-\alpha_n t}, \\
\mathrm{v}_\theta(r,\theta,t) &= \epsilon\omega_L R\left[\frac{A_n\omega_L}{\alpha_n}\left(\frac{r}{R}\right)^{n-1}\right. \\
&\quad \left. + \frac{B_n R}{n\,r}\left(j_n(kr) - \frac{kr\,j_{n+1}(kr)}{n+1}\right)\right]\frac{dP_n(\cos\theta)}{d\theta}e^{-\alpha_n t}, \\
\tau(\boldsymbol{r},t) &= \tau^{(n)}(\boldsymbol{r})\,e^{-\alpha_n t}, \quad \dot{\gamma}(\boldsymbol{r},t) = \dot{\gamma}^{(n)}(\boldsymbol{r})e^{-\alpha_n t}, \\
\omega_L &= \sqrt{\frac{\sigma n(n-1)(n+2)}{\rho R^3}}\,, \quad k = \sqrt{\frac{\rho\alpha_n}{\mu_{eff}}}
\end{aligned} \tag{S5}$$

where $\boldsymbol{r}$ is the position vector, ω_L the Lamb frequency, k the wave number, ρ the density of the liquid, j_n the spherical Bessel function of order n, and A_n and B_n unknown coefficients. The substitution of Eq. (S5) into the continuity and Navier-Stokes equations leads to a linear system of algebraic equations in A_n, B_n, C_n (see Ref. 2) from which the following characteristic equation is obtained:

$$\begin{aligned}
&\left(1+\frac{1}{x^2}\right)z^2 - 2\,Q(z)\left[\left(1+\frac{1}{x^2}\right) - \frac{2n(n-1)(n+2)}{z^2}\right] = 2(n-1)(2n+1), \\
&x = \frac{\alpha_n}{\omega_L} = \frac{\delta + i\omega}{\omega_L}, \quad Q(z) = \frac{z j_{n+1}(z)}{j_n(z)}, \quad z = kR
\end{aligned} \tag{S6}$$

S1.1 Low viscosity limit

For the limiting case when the effective viscosity is low ($z \to \infty$), we obtain simple formulae for viscosity μ and elastic modulus G:

$$\mu = \frac{\rho\delta R^2}{(n-1)(2n+1)}, \qquad G = \frac{(\omega^2 - \omega_L^2)\rho R^2}{2(n-1)(2n+1)}. \tag{S7}$$

In the case of quadrupole oscillation (n =2), Eq. (S7) is reduced to Eq. (4) in the main text.

S1.2 High viscosity limit

For the limiting case when the effective viscosity is high, the viscosity and elastic modulus are of the form

$$\mu = \frac{2F_1\delta\rho R^2}{F_2}, \qquad G = \frac{\rho R^2(F_1\omega^2 - \omega_l^2 + F_1\delta^2)}{F_2}$$

$$F_1 = \frac{12n^3 + 40n^2 + 38n + 15}{(2n+1)(2n+3)(2n+5)} \ \ and\ F_2 = \frac{4n^3 + 4n^2 - 2n - 6}{2n+1} \tag{S8}$$

For quadrupole oscillations (n = 2), F_1 = 16.524 and F_2 = 7.6 and

$$\mu = 4.348\ \delta\rho R^2, \quad G = \rho R^2(2.174\omega^2 - 0.132\omega_L^2 + 2.174\delta^2) \tag{S9}$$

S1.3 Forced oscillation of a viscoelastic drop

To derive the equation for forced oscillation, the surface profile of the drop [Eq. (S3)] and the boundary conditions [Eq. (S2)] were substituted into the radial component of the Navier-Stokes equations at the drop surface. Using the derivative of the pressure outside the drop as the forcing term, we then got the following driven harmonic oscillator equation:

$$\frac{d^2\ C_n(\mathrm{t})}{d\ t^2} + \frac{8\mu_{eff}}{\rho R^2}\left(\frac{dC_n(t)}{dt}\right) + \omega_L^2 C_n(t) \ = \frac{A\sin(\omega t)}{\rho R^3} \tag{S10}$$

The steady state solution for Eq. (S10) is

$$C_n(t) \ = \frac{A\sin(\omega t + \phi)}{\sqrt{\left(\rho R^3(\omega_L^2 - \omega^2)\right)^2 + \left(8\omega R\mu_{eff}\right)^2}}, \tag{S11}$$

$$\phi = \tan^{-1}\left[\frac{8\mu_{eff}\omega}{\rho R^2(\omega^2 - \omega_L^2\ )}\right],$$

where ϕ is the phase shift.

Section S2. Peak frequency for medical viscosity standard fluids

Figure S1 shows that the normalized peak frequency changed insignificantly between MVS fluids with viscosity from 4.0 to 6.0 cP. There was a slight, insignificant increase in the frequency for lower viscosity MVS fluids due to the multiple peaks in the AFR curve. Thus, ω_{peak} is not sensitive to fluid viscosity.

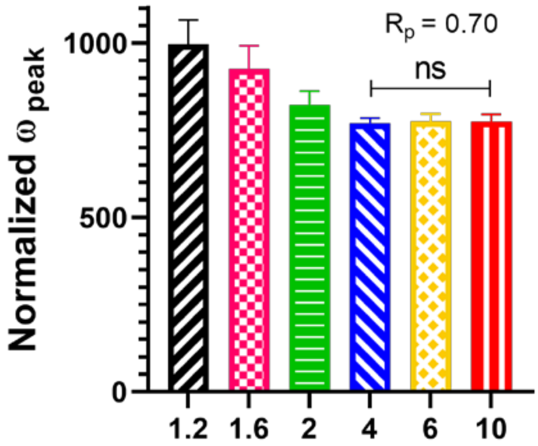

Fig. S1: Normalized peak frequency for medical viscosity standard fluids with viscosity 1.2cP (black), 1.6cP (pink), 2.0cP (green), 4.0cP (blue), 6.0cP (yellow) and 10cP (red). Sample size n = 9. "ns" stands for not significant.

References

1. M. A. Meyers and K. K. Chawla, *Mechanical Behavior of Materials*, Cambridge University Press, 2008.
2. D. B. Khismatullin and A. Nadim, *Physical Review E*, **63**, art. no.-061508, 2001.
3. R. B. Bird, R. C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids*, John Willey & Sons, 1987.