HIGH-FREQUENCY LINEAR VISCOSITY OF EMULSIONS COMPOSED OF TWO VISCOELASTIC FLUIDS

J. Blawzdziewicz*, E. Wajnryb**

*Department of Mechanical Engineering, Yale University, P.O. Box 208286, New Haven, CT 06520-8286

**Polish Academy of Sciences, IPPT PAN, Swietokrzyska 21, PL 00-049 Warszawa, Poland

Summary

The high-frequency linear response to an oscillatory flow is studied for an emulsion of viscoelastic droplets suspended in another viscoelastic fluid. Our analysis applies when the frequency of the imposed flow is much higher than the inverse capillary-relaxation time of the drops. However, the imposed frequency can be comparable to the inverse timescales associated with the response of the component fluids. In our approach, the complex, frequency-dependent effective viscosity of the emulsion is described using the Bergman spectral representation. It allows us to characterize the response of the system in the complex domain by a single real function, i.e. the spectral density. Moreover, the spectral representation enables construction of rapidly converging continued-fraction approximations. We find that the emulsion response is accurately described by several coefficients of the expansion. Numerical results for the spectrum and the continued-fraction coefficients are presented at different volume fractions for emulsions of randomly distributed drops.

Introduction

We consider high-frequency linear viscoelastic response of an emulsion composed of viscoelastic drops suspended in another viscoelastic fluid. Typical examples of such systems include polymer blends and emulsions of polymeric solutions. Due to importance of viscoelastic fluids in chemical, pharmacological, and food industries, viscoelastic emulsion properties have been intensively studied theoretically and experimentally. For moderate drop volume fractions and viscosity ratios between the drop and continuous fluids simple analytical expressions for the effective viscosity of an emulsion have been developed. However, such approximations are inaccurate at high volume fractions of highly viscous drops.

In this work we focus on the frequency regime where the timescale ω^{-1} associated with the frequency ω of the imposed flow is much smaller than the drop capillary relaxation time $\tau_\mathrm{d} = \eta a/\sigma$,

$$\omega\tau_\mathrm{d} \gg 1, \qquad (1)$$

Here a is drop radius, σ is the interfacial tension, and η is the average viscosity of the fluids. The imposed timescale ω^{-1} may, however, be comparable to the internal relaxation timescales characterizing the component fluids. In this frequency regime the effect of the interfacial tension on drop dynamics can be neglected; yet, the system has nontrivial viscoelastic response due to the viscoelasticity of the component fluids. The drop deformation is assumed to be small at all times, and the response of the component fluids is linear.

Under the high-frequency assumption (1) and small-deformation conditions the drops behave as nearly-spherical viscoelastic fluid blobs with complex viscosity ratio $\lambda(\omega) = \eta^\mathrm{in}(\omega)/\eta^\mathrm{out}(\omega)$, where $\eta^\mathrm{in}(\omega)$ and $\eta^\mathrm{out}(\omega)$ are complex, frequency-dependent viscosities of the drop and continuous-phase fluids. Due to the long capillary relaxation time (1) the drops passively react to the imposed flow, and the dependence of η^eff on ω stems entirely from the viscoelasticity of the component fluids, i.e.,

$$\eta^\mathrm{eff}(\omega) = \eta^\mathrm{out}(\omega)\bar{\eta}^\mathrm{eff}(\lambda(\omega)). \qquad (2)$$

Bergman representation

The structure of the function $\bar{\eta}^\mathrm{eff}(\lambda)$ is analyzed using the Bergman spectral representation [1]. The representation, corresponding to a pole expansion of the linear operator describing propagation of the microscopic field in the system, has the form

$$\chi_\mathrm{S} = \phi \int_0^1 \frac{g(q)}{q-s}\,\mathrm{d}q, \qquad (3)$$

where $\chi_\mathrm{S} = (\bar{\eta}^\mathrm{eff} - 1)/(\bar{\eta}^\mathrm{eff} + \frac{3}{2})$ is a normalized susceptibility of a spherical emulsion sample subject to an external flow, and ϕ is the volume fraction of drops. The spectral density $g(q)$ is a real function that depends only on the microscopic geometry (the distribution of the drops for the system considered herein), but is independent of the viscosities of the component fluids. The viscosity ratio in the above expression enters only through the Bergman variable

$$s = 1/(1-\lambda). \qquad (4)$$

Thus the complex effective viscosity (2) is represented in equation (4) via a real function of real variable $g(q)$.

We have derived explicit expressions for the function $g(q)$ in terms of the spectrum of the operators characterizing hydrodynamic interactions of spherical drops. The expressions have been evaluated numerically for different volume fractions of randomly distributed drops. Sample results of our calculations are presented in figure 1.

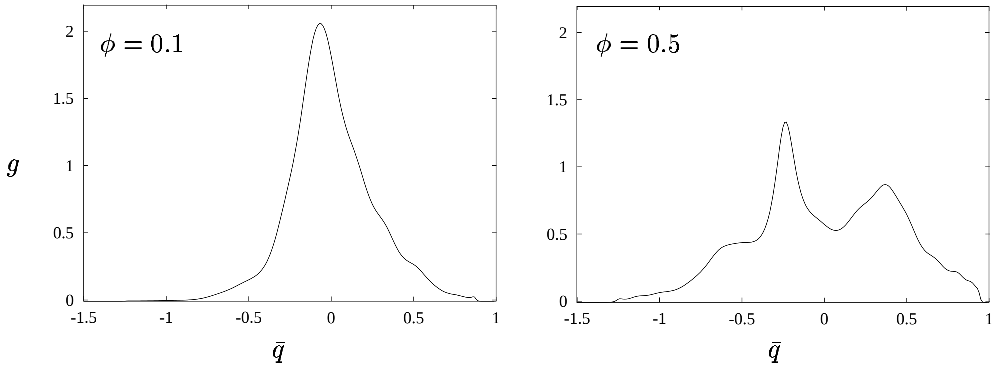

Figure 1. Spectral density g versus $\bar{q} = 1 - \frac{5}{2}q$ for two volume fractions ϕ (as labeled).

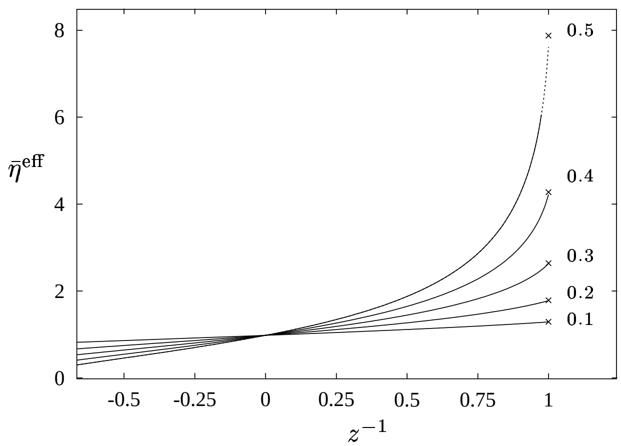

Figure 2. High frequency effective viscosity $\bar{\eta}^{\text{eff}}$ versus $z^{-1} = (\lambda - 1)/(\lambda + \frac{3}{2})$ for different volume fractions ϕ (as labeled). Solid lines correspond to the parameter region where results are accurate within 1 %. Crosses denote results for hard spheres ($\lambda = \infty$), obtained using a different method.

Continued-fraction expansion

The spectral representation not only reduces the amount of information needed to characterize linear response of the system, but it can also be used to construct a set of rapidly converging Padé approximants that form a sequence of increasingly tight rigorous bounds. Following the method used by [2], the Padé approximants are obtained here in a continued-fraction form

$$\tfrac{5}{2}\chi_{\mathrm{S}} = \cfrac{1}{s + \frac{2}{5} - \cfrac{a_1}{s + b_2 - \ldots}} \tag{5}$$

with the expansion coefficients evaluated numerically. Only several levels of the expansion (5) are needed to obtain the effective emulsion viscosity with high accuracy. Our results for the effective viscosity of an emulsion with real values of the viscosity ratio are shown in figure 2.

Conclusions

Using Bergman spectral representation, we have developed a complete description of high-frequency effective viscosity for an emulsion of viscoelastic fluids. We have evaluated numerically the spectral density and the coefficients of the continued-fraction expansion. With only several expansion levels, the continued fraction provides a very accurate analytical representation of the effective viscosity. The work is underway to extend this approach to the frequency regime $\omega\tau_{\mathrm{d}} \gg 1$, where the interfacial-tension effects are important.

References

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[2] B. Cichocki and B. U. Felderhof. Periodic fundamental solution of the linear Navier-Stokes equations. *Physica A*,159:19-27,1989

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