Measuring Transient Extensional Properties in Complex Microstructured Fluids using a Composite Harmonic Exponential Waveform (CHEW)

L.A. Kroo[1], P.T. Underhill[2], M.F. Rizzi[3], R.A. Nicholson[3] and G.H. McKinley[1]

[1] Dept. of Mechanical Eng., MIT, 77 Massachusetts Ave, Cambridge, MA 02139, USA (kroo@mit.edu)

[2] Dept. of Chemical and Biological Eng., Rensselaer Polytechnic Institute, 110 8th St, Troy, NY 12180, USA

[3] Research Division, Motif FoodWorks, Inc., 27 Drydock Ave, Boston, MA 02210, USA

Imposing complex strain histories on a material microstructure can result in a specific sequence of stress states that accurately emulates entirely different modalities of loading. Specifically, it was suggested by Doshi & Dealy (1987)[1] and later demonstrated by Kwan et al. (2001)[2] that an exponential shear strain will move the microstructure of a polymeric liquid through a similar sequence of stress states as a planar extensional flow. Fundamentally, this is enabled by the mechanical coupling between the normal and shear stresses that develop in complex fluids at large strains. Here we develop a *periodic* version of this concept, to probe dynamic extensional properties of complex fluids, using a conventional shear rheometer. The strain history is given by the following function: $\gamma(t) = A\sinh(\alpha\sin(\omega t))$, where A controls the maximum amplitude of the imposed strain and is normalized such that $A = \gamma_{max}/\sinh(\alpha)$, ω is the frequency of the cyclic deformation and the flow type parameter α effectively tunes the signal between a weak sinusoidal ($\alpha \ll 1$) deformation (e.g. SAOS with $\gamma_{max} << 1$, or LAOS for $\gamma_{max} \geq 1$) and a strong ($\alpha \gg 1$) deformation with exponential character. A representative Composite Harmonic Exponential Waveform (or CHEW strain history) is shown in Fig 1a, for α=10.

The benefits of expanding such a protocol to the periodic domain are two-fold: First, we can study the time-evolution of both shear and extensional properties of a complex fluid analogous to the convergence of medium- or large-amplitude oscillatory deformations into a limit cycle over the course of cyclic loading. Second, by measuring the shear stress and normal stress difference simultaneously we can also extract time-averaged extensional properties without specialized rheometric hardware (e.g. CABER).

We demonstrate this method experimentally on a viscous fluid with no elasticity, and a canonical viscoelastic fluid (3% wt PIB solution) as shown in Fig. 1b. Using CHEW and a standard torsional rheometer (Ares G2, TA Instruments), we can compute an appropriate periodic extensional viscosity function[3] ($\eta_{\text{ext}} = \Delta\sigma/(\alpha\omega)$) from the time-evolving principal normal stress difference (see: Fig 1c).

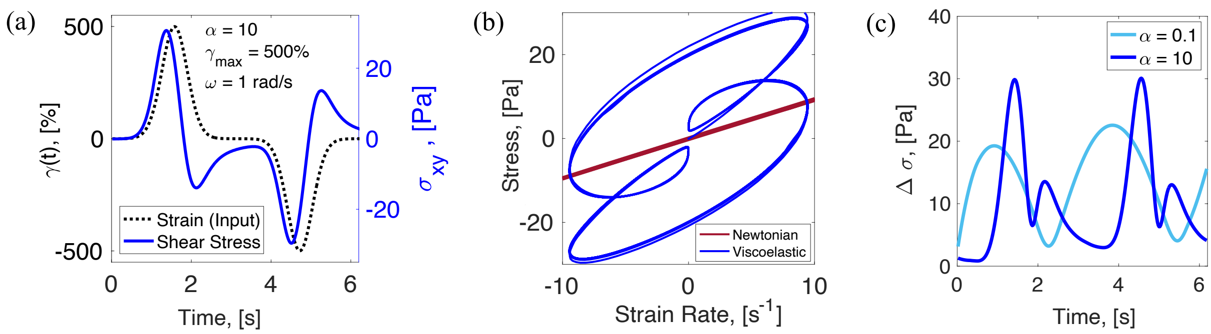

Figure 1: (a) A Composite Harmonic Exponential Waveform (black dotted) is shown in the limit of periodic exponential shear. The resulting shear stress response for a viscoelastic fluid (3 wt% PIB) is shown in blue. (b) Stress vs. strain rate for a Newtonian vs. viscoelastic fluid at α = 10. The linearity of the Newtonian response is evident and the maximum stress scales with the deformation rate $\dot{\gamma}_0 = A\alpha\omega$ (c) Evolution in the principal stress difference, $\Delta\sigma(t) = \sqrt{N1(t)^2 + 4\,\sigma_{xy}(t)^2}$ for a viscoelastic fluid over one cycle at $\alpha = 0.1$ vs $\alpha = 10$.

Additionally, to study the long-term adaptive effects (arising for example from thixotropic effects, or the Payne or Mullins effect in a complex fluid), we have constructed a nonlinear theoretical model to interpret the material response arising from CHEW. Finally, we use the CHEW protocol to measure the rheological response of multiphase semi-solid food materials and how they evolve with deformation time.

[1] Doshi, S. R., and J. M. Dealy. *Journal of Rheology* **31.7** (1987)

[2] T. Kwan, N. Woo, and ESG Shaqfeh. *Journal of Rheology* **45.2** (2001)

[3] M. Padmanabhan. *Journal of Food Engineering* **25.3** (1995)