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## Non-Newtonian viscous oscillating free surface jets, and a new strain-rate dependent viscosity form for flows experiencing low strain rates

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**Abstract** A model for oscillating free surface jet flow of a fluid from an elliptical orifice, together with experimental measurements, can be exploited to characterize the elongational viscosity of non-Newtonian inelastic fluids. The oscillating jet flow is predominantly elongational, with a small strain that oscillates rapidly between large and zero strain rates. We find that to reproduce the experimentally observed steady oscillating jet flow in model simulations, the assumed form of the non-Newtonian viscosity as a function of strain rate must have zero gradient, i.e., be Newtonian, at zero strain rate (a behavior exhibited, in general, by real inelastic fluids). We demonstrate that the Cross, Carreau, Prandtl-

Eyring, and Powell-Eyring forms, although they have finite viscosity at zero strain rate, have either nonzero or even unbounded gradient at zero, and hence are unable to model oscillating jet behavior. We propose a new non-Newtonian viscous form which has all of the desirable features of existing forms (high and low strain rate plateaus, with adjustable location and steepness of the transition) and the additional feature of Newtonian behavior at low strain rates.

**Key words** Oscillating free surface jet · Strain-rate thinning viscosity · Generalized Newtonian fluid · Non-Newtonian inelastic fluid · Rheometry · Extensional flows

### Introduction

When a fluid exits an elliptical orifice it establishes a jet with a free surface that is steady in time and oscillates in space (Fig. 1). Surface tension is the restoring force and viscosity is the damping; in broad strokes, the wavelength of the oscillating free surface is indicative of the surface tension of the fluid in the ambient atmosphere, and the decay of the oscillation indicates the fluid's viscosity. Lord Rayleigh (1879) developed a relation between surface tension and the wavelength of oscillation for an inviscid jet in the absence of gravity which, when combined with experimental measurements of oscillating jets, determines the surface tension of the fluid. Since then oscillating free surface jets have been used to measure surface tension; the advantage of this

technique is that, because the surface of the jet is newly created, the technique measures dynamic surface tension, i.e., tension as a rapidly changing function of time for newly formed surfaces. Pedersen (1907), Bohr (1909), Hansen et al. (1958), Defay and Hommelen (1958), and Thomas and Potter (1975) each present improvements to Rayleigh's relation. These algebraic equations for surface tension in terms of the measured wavelength and amplitude of the free surface oscillation neglect gravity, are based on a small departure of the oscillating profile from a circular cylinder, and model the fluid as Newtonian viscous. In previous work (Bechtel 1989; Bechtel et al. 1995, 1998) we derive models for a finite departure from equilibrium which account for gravity; these models take the form of nonlinear integro-differential equations for the free surface profile. In

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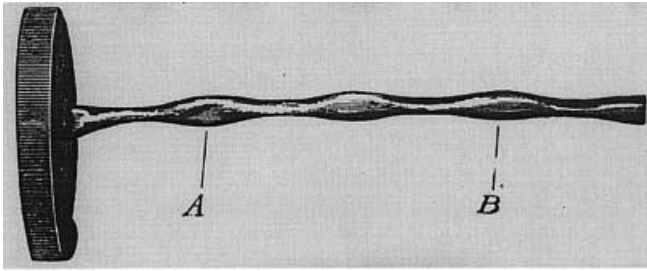


Fig. 1 The oscillating jet phenomenon; from Lord Rayleigh (1890)

Bechtel et al. (1995) we employ the inverse formulations of these models, in conjunction with experiments consistent with the model assumptions, to determine Newtonian viscosity as well as surface tension.

The model equations employed in all of the inverse techniques, except Bechtel et al. (1995), assume a constant surface tension and constant viscosity within the wavelength over which the measurements are taken. Bechtel et al. (1995) generalize the model to allow for variable surface tension and a non-Newtonian viscous fluid (although the non-Newtonian capability is not exploited in that paper). In Bechtel et al. (1998) we investigate the signature in Newtonian oscillating jet behavior of surface tension which changes within a wavelength and on submillisecond timescales. Here we investigate the effect of *non-Newtonian, strain-rate-dependent viscosity* on oscillating jet behavior.

In the investigation we have discovered that existing forms of strain-rate dependent viscosity (Cross, Carreau, Prandtl-Eyring, and Powell-Eyring) all exhibit anomalous non-zero gradients at low strain rates, so that they are unable to capture the Newtonian plateau exhibited in general in real inelastic fluids at low strain rates: Analysis reveals that the viscosity gradient at zero strain rate is unbounded for the Cross form, and finite but nonzero for the Carreau, Prandtl-Eyring, and Powell-Eyring forms.

In physical experiments, slowly varying (i.e., slender) oscillating jet flows of strain-rate-thinning non-Newtonian viscous fluids are readily observed, so the 3-D free surface boundary value problem should have solutions which are slowly varying. The kinematics of oscillating jet flow are such that if the effect of gravity is negligible (as is often the case), the strain rate is zero at the extremes of each free-surface oscillation. When the Cross, Carreau, Prandtl-Eyring, and Powell-Eyring forms are employed in the slender jet equations, they each fail at these locations of zero strain rate because of their non-zero gradients there, erroneously predicting the development of small length scales and subsequent jet breakup. The discovery that the slowly varying asymptotics can produce no consistent solution indicates a failure of the constitutive model for the fluid: the mathematical model has incorrect assumptions, and the only empirical ad hoc model feature is the viscosity form.

Our observations have an important ramification in the modeling of non-Newtonian viscous fluids in general. The oscillating jet flow serves as a test case. If a proposed viscosity form cannot model the oscillating jet flow, it is suspect to be applied to any flow that passes through low strain rates. An acceptable non-Newtonian viscous constitutive form must yield a solution for the oscillating jet problem which is slowly varying and stable when the fluid being modeled is observed to undergo a stable, slowly varying oscillating jet flow; to do this the form must possess, in addition to a bounded zero strain rate viscosity, a sufficiently small gradient at zero strain rate, i.e., it should be effectively Newtonian at low strain rates.

Motivated by this inadequacy of existing characterizations of non-Newtonian inelastic fluids, we present a new viscosity form, called the Zhou form, that has a zero strain rate viscosity gradient which is identically zero, i.e., it is Newtonian at low strain rates. We find that the Zhou form, unlike the other forms, leads to slender oscillating jet solutions.

### The governing equations for a steady viscous oscillating jet

In Bechtel et al. (1995) time-dependent governing equations for an oscillating slender jet of a viscous fluid are derived allowing for variable viscosity and surface tension. We recall only essential features of this derivation, which reveal why the various strain-rate dependent viscosity forms are inconsistent with slender oscillating jet behavior.

The fluid is assumed incompressible, and flows in the direction of gravity. We define Cartesian coordinates such that the  $e_3$  direction coincides with the centerline of the jet, and assume the jet cross section is elliptical for several oscillations, so that the free surface is

$$\frac{x_1^2}{\Phi_1^2(x_3, t)} + \frac{x_2^2}{\Phi_2^2(x_3, t)} - 1 = 0 \quad (1)$$

The fluid we model is viscous (not viscoelastic), with a strain-rate-dependent viscosity. Since the oscillating jet flow is predominantly elongational, the appropriate viscosity is elongational viscosity, which we label  $\eta$ ; if the fluid is Newtonian,  $\eta$  is constant and three times the shear viscosity. Here the viscosity is variable, an explicit function of the second invariant<sup>1</sup> of  $\mathbf{D}$ :

$$\hat{\mathbf{T}} = \frac{2}{3}\eta(\Pi_D)\mathbf{D}, \quad \Pi_D = \frac{1}{2} \left[ \text{tr}(\mathbf{D}^2) - (\text{tr}\mathbf{D})^2 \right], \quad (2)$$

where  $\hat{\mathbf{T}}$  is the determinate part of the Cauchy stress tensor  $\hat{\mathbf{T}} - p\mathbf{I}$  and  $\mathbf{D}$  is the symmetric part of the velocity gradient. For an incompressible fluid ( $\text{tr}\mathbf{D} = 0$ ),  $\Pi_D$  is

<sup>1</sup>The definition (2)<sub>2</sub> is not universal, e.g., Bird and Armstrong (1987) define their second invariant  $\Pi = 4\text{tr}(\mathbf{D}^2)$ , so that  $\Pi = 8\Pi_D$ .

non-negative. The customary definition of strain rate magnitude  $\dot{\gamma}$  is

$$\dot{\gamma} = 2\sqrt{\Pi_D} . \quad (3)$$

In order to deduce the dominant physical effects and exploit the slenderness (or, equivalently, slow variation) of the jet, the problem is non-dimensionalized through adoption of characteristic scales  $r_0$ ,  $z_0$ ,  $f_0$ ,  $v_0$  for transverse length, axial length, force, and axial velocity, respectively. The dimensionless forms  $x$ ,  $y$ ,  $z$ ,  $\tau$ ,  $\phi_1$ ,  $\phi_2$  of the coordinates  $x_1$ ,  $x_2$ ,  $x_3$ ,  $t$  and evolving semi-axes  $\Phi_1$ ,  $\Phi_2$  of the elliptical jet cross section are introduced through

$$x_1 = r_0x, \quad x_2 = r_0y, \quad x_3 = z_0z, \quad t = \frac{z_0}{v_0}\tau, \quad (4)$$

$$\Phi_1(x_3, t) = r_0\phi_1(z, \tau); \quad \Phi_2(x_3, t) = r_0\phi_2(z, \tau) .$$

The small parameter in the slender jet theory is the slenderness ratio  $\varepsilon = \frac{r_0}{z_0} \ll 1$ . For an oscillating jet  $\varepsilon$  is roughly the ratio of the mean radius of the jet cross section to the wavelength of the oscillation, typically 0.1. The velocity field within the oscillating jet is

$$\mathbf{v} = v_0 \{ [\varepsilon x \zeta_1(z, \tau) + O(\varepsilon^3)] \mathbf{e}_1 + [\varepsilon y \zeta_2(z, \tau) + O(\varepsilon_3)] \mathbf{e}_2 + (v(z, \tau) + O(\varepsilon_2)) \mathbf{e}_3 \} , \quad (5)$$

where  $\zeta_1$ ,  $\zeta_2$ ,  $v$  are dimensionless  $O(1)$  functions; note that the flow is elongational. For this flow the second invariant  $(2)_2$  is

$$\Pi_D = \frac{1}{2t_0^2} [\zeta_1^2 + \zeta_2^2 + v_z^2 + O(\varepsilon^2)] , \quad (6)$$

where “ ${}_z$ ” denotes differentiation with respect to the dimensionless axial coordinate  $z$ . To leading order in the slenderness ratio,  $\Pi_D$ , and hence the viscosity, are functions only of the axial coordinate and time.

To the fixed observer of an oscillating jet experiment, the free surface profile does not change; given the focus of this paper on variable viscosity, we restrict ourselves to constant surface tension. Therefore we recall the form of the governing non-dimensional equations for the oscillating jet which is steady with constant surface tension:

$$\begin{aligned} \zeta_1 &= \frac{v\phi_{1,z}}{\phi_1}, \quad \zeta_2 = \frac{v\phi_{2,z}}{\phi_2}, \quad v\phi_1\phi_2 = 1, \\ BP - 2Z\phi_1\phi_2\zeta_1 + \frac{\phi_1\phi_2}{W}K_c + \varepsilon^2 \frac{\phi_1^3\phi_2}{4} (Z_{,z}\zeta_{1,z} + Z\zeta_{1,zz}) \\ &= \frac{\phi_1^3\phi_2}{4} (v\zeta_{1,z} + \zeta_1^2), \\ BP - 2Z\phi_1\phi_2\zeta_2 + \frac{\phi_1\phi_2}{W}K_s + \varepsilon^2 \frac{\phi_1\phi_2^3}{4} (Z_{,z}\zeta_{2,z} + Z\zeta_{2,zz}) \\ &= \frac{\phi_2^3\phi_1}{4} (v\zeta_{2,z} + \zeta_2^2), \\ \varepsilon^2 \frac{1}{W} (\phi_1\phi_{2,z}K_s + \phi_2\phi_{1,z}K_c) + \varepsilon^2 (2\phi_1\phi_2Z_{,z}v_z \\ &+ 2\phi_1\phi_2Zv_{,zz} - BP) + \frac{1}{F}\phi_1\phi_2 = vv_{,z}\phi_1\phi_2 \end{aligned} \quad (7)$$

where

$$\begin{aligned} B &= \frac{z_0^2 f_0}{\rho v_0^2 r_0^4}, \quad \frac{1}{W} = \frac{\sigma z_0^2}{\rho v_0^2 r_0^3}, \quad \frac{1}{F} = \frac{gz_0}{v_0^2}, \quad Z(z) = \frac{\eta(z)z_0}{3\rho r_0^2 v_0}, \\ Z_{,z}(z) &= \frac{\eta_{,z}(z)z_0}{3\rho r_0^2 v_0}, \\ P &= \frac{1}{\pi f_0} \iint (p - p_a) dx_1 dx_2, \\ K_c &= \frac{1}{\pi} \int_0^{2\pi} \tilde{\kappa} \cos^2 \theta d\theta \\ &= -\frac{\phi_1\phi_2}{\pi} \int_0^{2\pi} \frac{\cos^2 \theta}{(\phi_1^2 \sin^2 \theta + \phi_2^2 \cos^2 \theta)^{3/2}} d\theta + O(\varepsilon^2), \\ K_s &= \frac{1}{\pi} \int_0^{2\pi} \tilde{\kappa} \sin^2 \theta d\theta \\ &= -\frac{\phi_1\phi_2}{\pi} \int_0^{2\pi} \frac{\sin^2 \theta}{(\phi_1^2 \sin^2 \theta + \phi_2^2 \cos^2 \theta)^{3/2}} d\theta + O(\varepsilon^2) ; \end{aligned} \quad (8)$$

$\rho$ ,  $\sigma$ ,  $g$ ,  $p_a$  are the fluid density, surface tension, acceleration of gravity, and specified ambient pressure, assumed constant, and  $\tilde{\kappa}$  is the dimensionless curvature of the surface.

Equations (7)<sub>1-3</sub> are the leading order equations which follow from the kinematic free surface boundary condition and incompressibility. These equations are independent of the constitutive nature of the fluid and hold for all regimes of jet behavior. The constant of integration in the flow rule (7)<sub>3</sub> presupposes that the characteristic length  $r_0$  is the geometric mean of the principal radii of the elliptical cross section at  $z=0$  and the characteristic velocity  $v_0$  is the axial velocity at  $z=0$ .

Equations (7)<sub>4-6</sub> correspond to projections of conservation of momentum in the  $x$ ,  $y$ , and  $z$  directions, respectively, and are special to the non-Newtonian viscous constitutive assumption (Eqs. 2). We have retained the leading order terms within each of the viscosity ( $Z$  and  $Z_{,z}$ ), surface tension ( $W$ ), constraint pressure ( $B$ ), gravity ( $F$ ), and inertia (1) terms (if surface tension is non-constant,  $W_{,z}$  terms appear in Eqs. (7)<sub>4-6</sub>; see Bechtel et al. 1998). For a Newtonian fluid the dimensionless parameter  $Z_{,z}$  is identically zero. The magnitude of the full set of dimensionless parameters (8)<sub>1-5</sub> relative to the slenderness ratio  $\varepsilon$  indicates the relative balances of the competing physical effects in the free jet problem. In a particular experiment some subset of these effects will be dominant, and thus survive in the leading order equations. The jet oscillates if inertia, surface tension, and the constraint pressure are leading order in the transverse momentum equations (7)<sub>4,5</sub>; this condition translates to  $\frac{1}{W}$ ,  $B \approx O(1)$ . We set  $B=1$  by fixing the force scale  $f_0 = \frac{\rho r_0^4 v_0^2}{z_0}$ , after which three independent scales remain:  $r_0$ ,  $z_0$  (typically  $10r_0$  for slender jets), and  $v_0$ , which

depend on the experimental conditions and, together with  $\rho, \sigma$ , and  $g$ , completely specify the oscillating jet behavior.

The consequences of variable viscosity in the asymptotic balance of Eqs. (7)<sub>4-6</sub> can be deduced by studying three terms:

$$L_0 = 2Z\phi_1\phi_2|\zeta_1|, \quad L_1 = \frac{\varepsilon^2\phi_1^3\phi_2}{4}|Z_{,z}\zeta_{1,z} + \zeta_{1,zz}Z|, \\ L_2 = 2\varepsilon^2|Z_{,z}v_{,z} + Zv_{,zz} - P|. \quad (9)$$

$L_0$  is the leading order viscosity term and  $L_1$  the next order correction in the transverse momentum equations (7)<sub>4,5</sub>, and  $L_2$  is the leading order viscosity term in the axial projection, (7)<sub>6</sub>.  $L_0$  must be  $O(1)$  for oscillating viscous jets (if  $L_0$  is  $O(\varepsilon^2)$  or less the oscillating jet is inviscid, with no damping to leading order). The equations (7)<sub>4-6</sub> simplify to slender jet equations *only if* the correction terms  $L_1$  and  $L_2$  are  $O(\varepsilon^2)$ ; otherwise these terms compete in the leading order equations with the formally leading-order  $L_0$  term, thereby disordering the perturbation expansion.

To anticipate, in the analysis immediately following we show that for the power law, Cross, Carreau, Prandtl-Eyring, and Powell-Eyring forms giving viscosity  $\eta$  as a function of second invariant  $\Pi_D$ , the viscosity gradient  $\frac{d\eta}{d(\Pi_D)}$  is nonzero at  $\Pi_D = 0$ , i.e., at zero strain rate. This feature is critical in the oscillating jet characterization precisely because in that flow the strain rate oscillates about zero: we discover in the computations of the next section that, since  $\frac{d\eta}{d(\Pi_D)}(0) \neq 0$ , the dimensionless viscosity gradient  $Z_{,z}$  becomes too large at the axial locations of small strain rates for these standard forms, causing  $L_1$  to be huge. This implies that *either* the jet flow cannot remain slender *or* the empirical viscosity forms are problematic. Since there is no experimental evidence of an inability to perform oscillating slender jet experiments, we conclude these empirical viscosity forms have unphysical features for small strain rates.

We define the non-dimensional strain rate magnitude  $\tilde{\gamma}$  by

$$\tilde{\gamma} = \frac{z_0}{v_0} \sqrt{\Pi_D} = \frac{z_0}{2v_0} \dot{\gamma}. \quad (10)$$

The dimensionless form of the viscosity  $\eta(\Pi_D)$  is then

$$Z = Z(\tilde{\gamma}^2). \quad (11)$$

To leading order in the oscillating jet,

$$\tilde{\gamma} = \sqrt{\frac{1}{2}(\zeta_1^2 + \zeta_2^2 + v_z^2)}. \quad (12)$$

### Power law fluid

In the power law model (Bird et al. 1987) viscosity is described by the expression<sup>2</sup>

<sup>2</sup>In Bird et al. (1987),  $\eta = k\sqrt{\frac{l-1}{2}}$ , which is equivalent to Eq. (14), noting  $\Pi = 8\Pi_D$  from the previous footnote.

$$\eta = k(4\Pi_D)^{\frac{l-1}{2}}, \quad (13)$$

where the consistency index  $k$  and power law exponent  $l$  are positive constants. The fluid is strain-rate thinning for  $l < 1$ , Newtonian for  $l = 1$ , and strain-rate thickening for  $l > 1$ .

For the power law form the viscosity gradient is

$$\frac{d\eta}{d(\Pi_D)} = k(l-1)2^{l-2}(\Pi_D)^{\frac{l-3}{2}}. \quad (14)$$

The dimensionless forms of the viscosity (Eq. 13) and viscosity gradient (Eq. 14) are

$$Z = Z_P\tilde{\gamma}^{l-1}, \quad Z' \equiv \frac{dZ(\tilde{\gamma}^2)}{d\tilde{\gamma}^2} = \frac{(l-1)}{2}Z_P\tilde{\gamma}^{l-3}, \quad (15)$$

with the two dimensionless constants

$$l, \quad Z_P = \frac{kz_0}{3\rho r_0^2 v_0} \left(\frac{2v_0}{z_0}\right)^{l-1}. \quad (16)$$

For a strain-rate thinning fluid, as the strain rate goes to zero,

$$\eta(0) \rightarrow \infty, \quad \frac{d\eta}{d(\Pi_D)}(0) \rightarrow -\infty, \quad Z(0) \rightarrow \infty, \quad Z'(0) \rightarrow -\infty. \quad (17)$$

### Cross model

The predictions for a strain-rate thinning fluid of infinite viscosity when the strain rate is zero, and zero viscosity when the strain rate is large, are shortcomings of the power law model. Cross (1965)<sup>3</sup> proposed a model with two additional constants  $\eta_0$  and  $\eta_\infty$  that corrects these shortcomings:

$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + (4k^2\Pi_D)^{\frac{1-l}{2}}}. \quad (18)$$

If  $l < 1$  and  $\eta_0 > \eta_\infty > 0$ , the viscosity is strain-rate thinning;  $\eta_0$  and  $\eta_\infty$  are the limiting viscosities at zero and high strain rates, respectively. If  $l > 1$  and  $\eta_0 > \eta_\infty > 0$ , the fluid is strain-rate thickening, with  $\eta_0$  and  $\eta_\infty$  reversing roles to be the limiting viscosities at high and zero strain rates, respectively. The Cross form reduces to constant viscosity either by setting  $l = 1$  (resulting in constant viscosity  $\frac{\eta_0 + \eta_\infty}{2}$ ) or setting  $\eta_0 = \eta_\infty$ .

The viscosity gradient as a function of strain rate is

$$\frac{d\eta}{d(\Pi_D)} = (l-1) \frac{k^{l-1}}{2^l} \frac{(\eta_0 - \eta_\infty)}{(\Pi_D)^{\frac{1+l}{2}} \left[1 + (4k^2\Pi_D)^{\frac{1-l}{2}}\right]}. \quad (19)$$

<sup>3</sup>Cross gives  $\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + (\tau D)^m}$ , in which  $D = 2\sqrt{\Pi_D}$ . This is equivalent to Eq. (18) if  $\tau = k$ ,  $m = 1-l$ . In his original paper Cross fixes  $m = \frac{2}{3}$ , i.e.,  $l = \frac{1}{3}$ .

The dimensionless forms of the viscosity (Eq. 18) and viscosity gradient (Eq. 19) are

$$Z = Z_\infty + \frac{Z_0 - Z_\infty}{1 + Z_C \tilde{\gamma}^{1-l}},$$

$$Z' \equiv \frac{dZ(\tilde{\gamma}^2)}{d\tilde{\gamma}^2} = Z_C \frac{(l-1)(Z_0 - Z_\infty)}{2\tilde{\gamma}^{l+1}(1 + Z_C \tilde{\gamma}^{1-l})^2}, \quad (20)$$

where there are now four dimensionless constants,

$$l, Z_\infty = \frac{z_0}{3\rho r_0^2 v_0} \eta_\infty, Z_0 = \frac{z_0}{3\rho r_0^2 v_0} \eta_0, Z_C = \left(\frac{2kv_0}{z_0}\right)^{1-l}. \quad (21)$$

For a strain-rate thinning fluid, as the strain rate goes to zero the viscosity is bounded, *but the viscosity gradient is unbounded*,

$$\eta(0) = \eta_0, \frac{d\eta}{d(\Pi_D)}(0) \rightarrow -\infty, Z(0) = Z_0, Z'(0) \rightarrow \infty; \quad (22)$$

the Cross fluid is strongly non-Newtonian at low strain rates.

#### Carreau model

Another remedy for the power law's unbounded viscosity at zero strain-rate is the Carreau (1972) viscosity model<sup>4</sup>, which has been shown to have enough flexibility to fit several experimental viscosity curves (Bird et al. 1987; Macosko 1993):

$$\eta = \eta_\infty + (\eta_0 - \eta_\infty)(1 + 4\lambda^2 \Pi_D)^{\frac{l-1}{2}}, \quad (23)$$

where  $\lambda$  is a time constant. If  $l < 1$  and  $\eta_0 > \eta_\infty > 0$  this form models strain-rate thinning, whereas  $l < 1$  and  $0 < \eta_0 < \eta_\infty$  models strain-rate thickening. In both cases,  $\eta_0$  and  $\eta_\infty$  are the limiting viscosities at zero and high strain rates, respectively. The form reduces to Newtonian either by setting  $l=1$  (resulting in constant viscosity  $\eta_0$ ) or setting  $\eta_0 = \eta_\infty$ .

The viscosity gradient as a function of strain rate is

$$\frac{d\eta}{d(\Pi_D)} = 2\lambda^2(l-1)(\eta_0 - \eta_\infty)(1 + 4\lambda^2 \Pi_D)^{\frac{l-3}{2}}, \quad (24)$$

and the dimensionless viscosity and viscosity gradient are

$$Z = Z_\infty + (Z_0 - Z_\infty) \left[1 + (Z_C \tilde{\gamma})^2\right]^{\frac{l-1}{2}},$$

$$Z' = Z_C^2(l-1) \frac{(Z_0 - Z_\infty)}{2} \left[1 + (Z_C \tilde{\gamma})^2\right]^{\frac{l-3}{2}}, \quad (25)$$

where the four dimensionless constants are  $l$ , the  $Z_0$  and  $Z_\infty$  defined in Eq. (21)<sub>2,3</sub>, and

$$Z_{CY} = \frac{2\lambda v_0}{z_0}. \quad (26)$$

For this form, both the viscosity and viscosity gradient are finite at zero strain rate for strain-rate thinning fluids, *but the gradient is nonzero*:

$$\eta(0) = \eta_0, \frac{d\eta}{d(\Pi_D)}(0) = 2\lambda^2(l-1)(\eta_0 - \eta_\infty),$$

$$Z(0) = Z_0, Z'(0) = Z_{CY}^2(l-1) \frac{(Z_0 - Z_\infty)}{2}. \quad (27)$$

Again, this model predicts non-Newtonian behavior at low strain rates. The only way the Carreau form can model Newtonian behavior at low strain rates is if it models the fluid as Newtonian at all strain rates, since the viscosity gradient at zero strain rate is zero only if  $l=1$  or  $\eta_0 = \eta_\infty$ .

#### Prandtl-Eyring model

One of the first viscosity forms obtained by a molecular theory was the Eyring equation (Brodkey 1967). A modification of this form, the Prandtl-Eyring model (Skelland 1967) has been used to describe the rheology of strain-rate thinning fluids:

$$\eta(\Pi_D) = \frac{B\eta_0 \sinh^{-1}\left(\frac{\sqrt{\Pi_D}}{B}\right)}{\sqrt{\Pi_D}}, \quad (28)$$

where  $B$  is a constant. The limit when the strain rate is zero is  $\eta_0$ . Like the power law, this form can only predict inviscid behavior at high strain rates. The viscosity gradient as a function of strain rate is

$$\frac{d\eta}{d(\Pi_D)} = \frac{\eta_0 \sqrt{\Pi_D} - B \sqrt{1 + \frac{\Pi_D}{B^2}} \sinh^{-1}\left(\frac{\sqrt{\Pi_D}}{B}\right)}{2 \Pi_D^{3/2} \sqrt{1 + \frac{\Pi_D}{B^2}}}, \quad (29)$$

which is *finite but nonzero* at zero strain rate (as with the Carreau form):

$$\frac{d\eta}{d(\Pi_D)}(0) = -\frac{\eta_0}{6B^2}. \quad (30)$$

#### Powell-Eyring model

The Powell-Eyring model (Skelland 1967) adds a constant to the Prandtl-Eyring form:

$$\eta(\Pi_D) = \frac{B(\eta_0 - \eta_\infty) \sinh^{-1}\left(\frac{\sqrt{\Pi_D}}{B}\right)}{\sqrt{\Pi_D}} + \eta_\infty, \quad (31)$$

resulting in a finite limit  $\eta_\infty$  at high strain rates, but the viscosity gradient is unaltered from the Prandtl-Eyring model, with the same zero strain rate value given in Eq. (30).

<sup>4</sup>The Carreau-Yasuda model is given in Bird et al. (1987) in the form  $\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = [1 + (\lambda\dot{\gamma})^a]^{\frac{l-1}{a}}$  where  $\gamma = 2\sqrt{\Pi_D}$ . This model with the parameter  $a=2$ , referred to as the Carreau model, is equivalent to Eq. (23).

## Zhou model

We now propose a form which has a finite value of viscosity and a zero viscosity gradient at zero strain rate, representing for the first time a *non-Newtonian viscous fluid model which is truly Newtonian at low strain rates*:

$$\eta = \frac{\eta_0 + \eta_\infty}{2} + \left(\frac{\eta_0 - \eta_\infty}{2}\right) \frac{1 - f(\Pi_D)}{1 + f(\Pi_D)},$$

$$f(\Pi_D) = \left(\frac{\Pi_D}{A}\right)^{\frac{1}{\alpha}} \exp\left\{B \left[1 - \left(\frac{A}{\Pi_D}\right)^{\frac{1}{\alpha}}\right]\right\}. \quad (32)$$

Specifying  $A > 0$ ,  $B > 0$ ,  $\frac{1}{\alpha} > 0$ ,  $\eta_0 > \eta_\infty > 0$  models a strain-rate thinning fluid. The constants  $\eta_0$  and  $\eta_\infty$  are the limiting viscosities at zero and high strain rates, respectively,  $A$  is the strain rate at the center of the transition from the low strain rate viscosity plateau  $\eta_0$  to the high strain rate plateau  $\eta_\infty$ , and  $\alpha$  controls the steepness of this drop. *The constant  $B$  is proportional to the range of strain rates over which the fluid is strictly Newtonian*, i.e., the viscosity is strictly constant.

The viscosity gradient for this model is

$$\frac{d\eta}{d(\Pi_D)} = (\eta_\infty - \eta_0) \frac{f'(\Pi_D)}{[1 + f(\Pi_D)]^2},$$

$$f'(\Pi_D) = \frac{1}{\alpha} \left[ \frac{1}{A} \left(\frac{\Pi_D}{A}\right)^{\frac{1}{\alpha}-1} + \frac{B}{\Pi_D} \right] \exp\left\{B \left[1 - \left(\frac{A}{\Pi_D}\right)^{\frac{1}{\alpha}}\right]\right\}. \quad (33)$$

In dimensionless form the viscosity and viscosity gradient are

$$Z = \frac{Z_0 + Z_\infty}{2} + \frac{Z_0 - Z_\infty}{2} \frac{1 - (Z_H \tilde{\gamma}^2)^{\frac{1}{\alpha}} \exp\left\{B \left[1 - \left(\frac{1}{Z_H \tilde{\gamma}^2}\right)^{\frac{1}{\alpha}}\right]\right\}}{1 + (Z_H \tilde{\gamma}^2)^{\frac{1}{\alpha}} \exp\left\{B \left[1 - \left(\frac{1}{Z_H \tilde{\gamma}^2}\right)^{\frac{1}{\alpha}}\right]\right\}},$$

$$Z' = (Z_\infty - Z_0) \frac{\frac{1}{\alpha} \left[ \frac{1}{A} (Z_H \tilde{\gamma}^2)^{\frac{1}{\alpha}-1} + \frac{B}{\tilde{\gamma}^2} \right] \exp\left\{B \left[1 - (Z_H \tilde{\gamma}^2)^{\frac{1}{\alpha}}\right]\right\}}{\left[1 + (Z_H \tilde{\gamma}^2)^{\frac{1}{\alpha}} \exp\left\{B \left[1 - \left(\frac{1}{Z_H \tilde{\gamma}^2}\right)^{\frac{1}{\alpha}}\right]\right\}\right]^2}, \quad (34)$$

where there are now five dimensionless constants:  $\alpha$ ,  $B$ , the  $Z_0$  and  $Z_\infty$  defined in Eqs. (21)<sub>2,3</sub>, and

$$Z_H = \frac{z_0^2}{A v_0^2}. \quad (35)$$

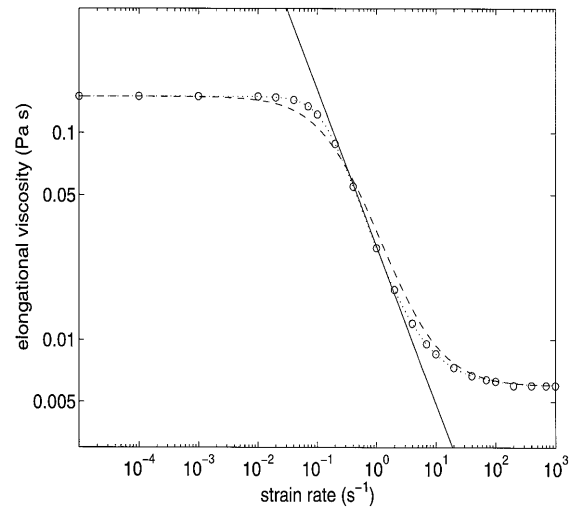
For this form the viscosity is finite and the viscosity gradient is zero at zero strain rate for strain-rate thinning fluids:

$$\eta(0) = \eta_0, \quad \frac{d\eta}{d(\Pi_D)}(0) = 0, \quad Z(0) = Z_0, \quad Z'(0) = 0. \quad (36)$$

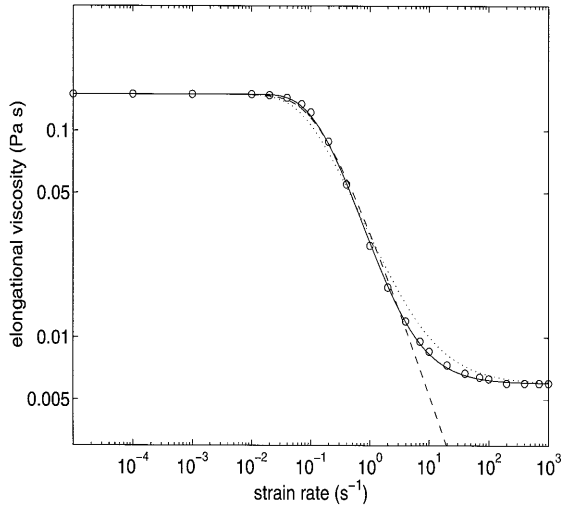
Of all the strain-rate thinning viscosity forms considered, we will find that because of its zero gradient at zero strain rate this form, and only this form, can model the oscillating jet flow.

## Example characterization of a conceptual fluid

To demonstrate the differences between the above six non-Newtonian viscosity forms, we fit all of the forms to the same set of elongational viscosity vs strain rate data points, the circles  $\circ$  in Figs 2, 3, and 4. For complex fluids elongational response cannot in general be inferred from easy-to-perform shear measurements (Moore and Pearson 1975; Baid and Metzner 1977; Chai and Yeow 1988; Ramanan et al. 1997), and it is a significant challenge to produce viscosity vs strain rate data points directly from elongational measurements of an inelastic fluid. We comment that the inverse formulation of the mathematical model presented here, together with experimental measurements of the oscillating jet free surface profile, can be used to produce data sets like the circles in Figs. 2, 3, and 4, but that remains for future work. For the demonstration purposes of this paper we have created a data set which exhibits strain-rate thinning. Explicitly, we create a hypothetical fluid with a low strain-rate Newtonian elongational viscosity plateau of 0.15 Pa s for strain-rates  $1 \times 10^{-5} \text{ s}^{-1}$ ,  $1 \times 10^{-4} \text{ s}^{-1}$ ,  $1 \times 10^{-3} \text{ s}^{-1}$ , and  $1 \times 10^{-2} \text{ s}^{-1}$ , and a high strain-rate Newtonian plateau of 0.006 Pa s for strain rates  $2 \times 10^2 \text{ s}^{-1}$ ,  $4 \times 10^2 \text{ s}^{-1}$ ,  $7 \times 10^2 \text{ s}^{-1}$ , and  $1 \times 10^3 \text{ s}^{-1}$ . The transition data points at the strain rates  $0.02 \text{ s}^{-1}$ ,  $0.04 \text{ s}^{-1}$ ,  $0.07 \text{ s}^{-1}$ ,  $0.1 \text{ s}^{-1}$ ,



**Fig. 2** Plot of log of viscosity vs log of strain rate  $\dot{\gamma}$  for strain-rate thinning viscosity forms fit to the experimental data ( $\circ$ ): power law (—), Cross (---), Carreau (···); coefficients in the forms are given in Table 1. The power law form predicts unbounded viscosity at low strain rates and a viscosity which approaches zero at high strain rates



**Fig. 3** Plot of log of viscosity vs log of strain rate  $\dot{\gamma}$  for strain-rate thinning viscosity forms fit to the experimental data (O): Prandtl-Eyring (---), Powell-Eyring (···), Zhou (—); coefficients in the forms are given in Table 1. The Prandtl-Eyring form predicts a viscosity which approaches zero at high strain rates

$0.2 \text{ s}^{-1}$ ,  $0.4 \text{ s}^{-1}$ ,  $0.7 \text{ s}^{-1}$ ,  $1 \text{ s}^{-1}$ ,  $2 \text{ s}^{-1}$ ,  $4 \text{ s}^{-1}$ ,  $7 \text{ s}^{-1}$ ,  $10 \text{ s}^{-1}$ ,  $20 \text{ s}^{-1}$ ,  $40 \text{ s}^{-1}$ ,  $70 \text{ s}^{-1}$ , and  $100 \text{ s}^{-1}$  are generated from the Carreau form (Eq. 23) with  $\eta_\infty = 0.006 \text{ Pa s}$ ,  $\eta_0 = 0.15 \text{ Pa s}$ ,  $\lambda = 7.57 \text{ s}$ , and  $l = 0.068$ .

We fit each viscosity form to the data as follows: the power law coefficients are obtained using a least squares fit of only the data between  $10^{-2} \text{ s}^{-1}$  and  $10^2 \text{ s}^{-1}$ , the coefficients in the Prandtl-Eyring form are obtained using a least squares fit of only the data between  $10^{-5} \text{ s}^{-1}$  and  $10^2 \text{ s}^{-1}$ , and the coefficients in the other four forms are obtained using a least squares fit of all data (even though we have used the Carreau form to generate the transition data, we fit it to all of the data, since as we will see the Carreau form is incapable of capturing the Newtonian low strain-rate plateau). As is customary, the fits are done on  $\log \eta$ . The resulting characterizations are in Table 1.

Figures 2 and 3 give the customary representation of the strain rate dependent viscosity, namely a plot of log of viscosity vs log of the strain rate  $\dot{\gamma} = 2\sqrt{\Pi_D}$ . The power law and Prandtl-Eyring forms are unable to capture the nonzero viscosity plateau at high strain rates. All of the forms except for the power law *appear in these log-log plots* to have a Newtonian plateau at low

strain rates. This appearance is deceptive: non-log plots of viscosity vs strain rate and viscosity gradient vs strain rate at low strain rates (Figs. 4 and 5, respectively) and the left column of Table 1 reveal that all but our Zhou form have negative viscosity gradient, rather than zero gradient, throughout the low strain rate “plateau.” As can be seen from Figs. 4 and 5, only our form is capable of modeling a fluid with constant viscosity over a range of low strain rates, the feature that proves to be critical to model oscillating jets. Note also from Fig. 3 that our Zhou form is capable of fitting the transition behavior predicted by the Carreau form, more so than the power law, Cross, Prandtl-Eyring, and Powell-Eyring forms.

We now apply the six constitutive forms to model oscillatory jet behavior.

### Viscous oscillating jets

We assume that the material properties and process conditions are such that  $W$  and  $Z$  are  $O(1)$  and  $Z'$  and  $\frac{1}{F}$  are at most  $O(\varepsilon^2)$ , so that the leading order equations from Eq. (7) are, in order:

$$\begin{aligned} \zeta_1 &= \frac{\phi_{1,z}}{\phi_1}, \quad \zeta_2 = \frac{\phi_{2,z}}{\phi_2}, \quad \phi_1 \phi_2 = 1, \\ P - 2Z\zeta_1 + \frac{1}{W}K_c &= \frac{\phi_1^2}{4}(\zeta_{1,z} + \zeta_1^2), \\ P - 2Z\zeta_2 + \frac{1}{W}K_s &= \frac{\phi_2^2}{4}(\zeta_{2,z} + \zeta_2^2), \quad v = 1. \end{aligned} \quad (37)$$

In this regime, pressure, surface tension, inertia, and viscosity balance in the transverse oscillation, and gravity, surface tension, viscosity, and viscosity gradients are too weak to accelerate the jet in the axial direction in this order of approximation. The system of Equations (37) reduces algebraically to a single integro-differential equation for the free surface profile  $\phi_1$ :

$$(1 + \phi_1^4)\phi_{1,zz} - \frac{2}{\phi_1}\phi_{1,z}^2 + \frac{4\phi_1^3}{W}(K_s - K_c) + 16Z\phi_1^2\phi_{1,z} = 0, \quad (38)$$

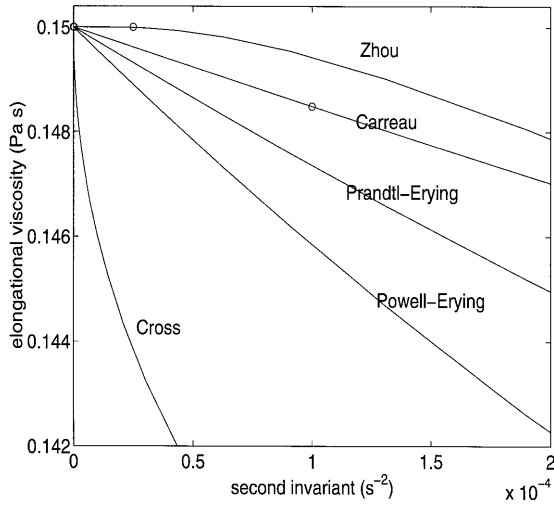
where

$$K_s - K_c = \frac{\phi_1^3}{\pi} \int_0^{2\pi} \frac{\cos^2 \theta - \sin^2 \theta}{(\cos^2 \theta + \phi_1^4 \sin^2 \theta)^{3/2}} d\theta.$$

**Table 1** Coefficients in the viscosity forms characterizing the conceptual fluid

Model	Coefficients	$\frac{d\eta}{d(\Pi_D)}(0)$
Power law	$k = 0.0282 \text{ Pa s}^{0.48}$ , $l = 0.48$	$-\infty$
Cross	$\eta_\infty = 0.15 \text{ Pa s}$ , $\eta_0 = 0.006 \text{ Pa s}$ , $k = 6.18 \text{ s}$ , $l = .11$	$-\infty$
Carreau	$\eta_\infty = 0.15 \text{ Pa s}$ , $\eta_0 = 0.006 \text{ Pa s}$ , $\lambda = 7.57 \text{ s}$ , $l = 0.068$	$-15.4 \text{ Pa s}^3$
Prandtl-Eyring	$\eta_0 = 0.15 \text{ Pa s}$ , $B = 0.0301 \text{ s}^{-1}$	$-27.7 \text{ Pa s}^3$
Powell-Eyring	$\eta_0 = 0.15 \text{ Pa s}$ , $B = 0.0231 \text{ s}^{-1}$ , $\eta_\infty = 0.006 \text{ Pa s}$	$-46.8 \text{ Pa s}^3$
Zhou	$\eta_\infty = 0.006 \text{ Pa s}$ , $\eta_0 = 0.15 \text{ Pa s}$ , $A = 1.4102 \times 10^{-2} \text{ s}^{-2}$ , $B = 0.25$ , $\alpha = 1.9345$	0





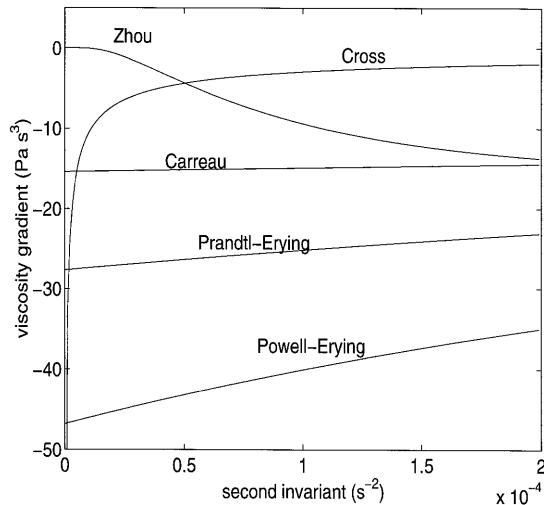
**Fig. 4** Non-log plot of viscosity vs second invariant  $\Pi_D$  for the strain-rate thinning viscosity models of Figs. 2 and 3. Experimental data ( $\circ$ ). Only the Zhou form models the Newtonian (constant viscosity) behavior of the fluid at low strain rates: the Cross form has unbounded slope at zero, the Carreau, Prandtl-Eyring, and Powell-Eyring forms have negative slope at zero, and the Zhou form has zero slope at zero

The correction terms defined in Eq. (9)<sub>2,3</sub> reduce to

$$L_1 = \frac{\varepsilon^2 \phi_1^4}{4} \left| 2(\tilde{\gamma}_{,z})^2 Z' + Z \tilde{\gamma}_{,zz} \right|, \tag{39}$$

$$L_2 = \varepsilon^2 \left| \frac{K_c}{W} - 2Z\tilde{\gamma} - \frac{\phi_1^2}{4} (\tilde{\gamma}_{,z} + \tilde{\gamma}^2) \right|;$$

recall that a solution of the slender jet (equation 38) is valid *only if* these terms remain small, i.e.,  $O(\varepsilon^2)$ .



**Fig. 5** Viscosity gradient vs second invariant  $\Pi_D$  for the strain-rate thinning viscosity models

To leading order the non-dimensional strain rate magnitude (Eq. 12) is

$$\tilde{\gamma} = \left| \frac{\phi_{1,z}}{\phi_1} \right|. \tag{40}$$

Note that in this regime the strain rate is zero at all locations where  $\phi_{1,z} = 0$ , i.e., at the maxima and minima of the free surface oscillation (recall  $\phi_1(z)$  is the evolving semi-axis of the elliptical cross section). Hence, in this regime of oscillating jet behavior the strain rate repeatedly passes through zero, twice in each wavelength.

**Power law fluid**

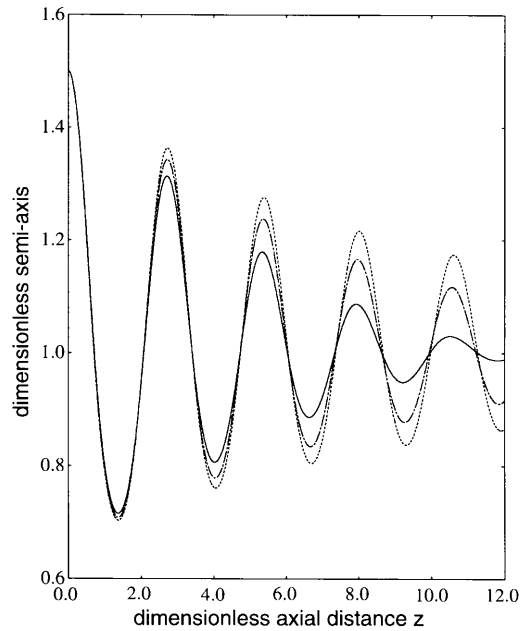
When the power law expression (15), for dimensionless viscosity  $Z$  is inserted into Eq. (38) we get

$$\begin{aligned} (1 + \phi_1^4) \phi_{1,zz} - \frac{2}{\phi_1} \phi_{1,z}^2 + \frac{4\phi_1^3}{W} (K_s - K_c) \\ + 16Z_P \phi_1^{3-l} \text{sign}(\phi_{1,z}) |\phi_{1,z}|^l = 0. \end{aligned} \tag{41}$$

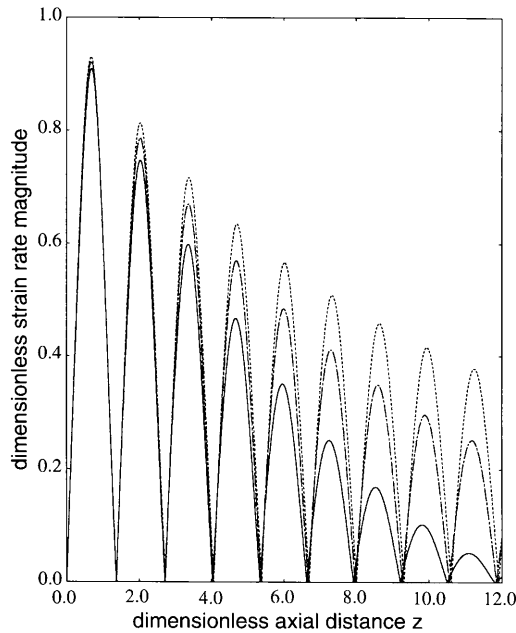
The solution of the dimensionless integro-differential equation (41) for the free surface profile  $\phi_1$  depends on initial conditions  $\phi_1(0)$  and  $\phi_{1,z}(0)$ , and three constants  $W$ ,  $Z_P$ , and  $l$ :

$$\phi_1 = \hat{\phi}_1(W, Z_P, l). \tag{42}$$

Figure 6 displays leading order solutions for strain-rate thinning ( $l=0.5$ ), Newtonian ( $l=1$ ), and strain-rate



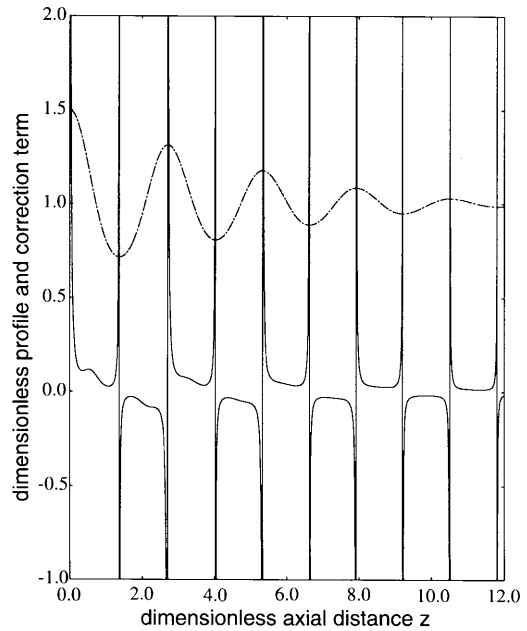
**Fig. 6** Free surface profiles  $\phi_1(z)$  for inviscid jets with rate-dependent viscosity given by the power law:  $\phi_1(0)=1.5$ ,  $\phi_{1,z}(0)=0$ ,  $W=1$ ,  $Z_P=0.032$ , with  $l=0.5$  (strain-rate thinning,  $\text{---}$ ),  $l=1$  (Newtonian,  $\text{- - -}$ ),  $l=1.5$  (strain-rate thickening,  $\text{...}$ )



**Fig. 7** Dimensionless strain rate magnitude  $\tilde{\gamma}$  for the solutions in Fig. 6. The strain rate is zero at all locations of extrema of the free surface oscillation

thickening ( $l=1.5$ ) fluids, holding  $W$  and  $Z_p$  fixed. Figure 7 displays the accompanying evolution of the strain rate magnitude; these simulations confirm that the strain rate repeatedly passes through zero at the locations of minima and maxima of the free surface oscillation.

We now check if the correction terms  $L_1$  and  $L_2$  remain  $O(\varepsilon^2)$  everywhere, as must be the case if the solution of Eq. (41) is to be consistent with the assumptions inherent in Eq. (41).  $L_2$  behaves properly. However  $L_1$  is in general unbounded at locations of zero strain rate for all  $0 < l < 1$  (all strain-rate thinning fluids) and  $1 < l < 3$  (a range of strain-rate thickening fluids). From Eq. (16), the viscosity gradient  $Z'$  goes to infinity for the power law fluid as strain rate  $\tilde{\gamma}$  goes to zero if  $l < 3$ ; therefore the product  $(\tilde{\gamma}_{,z})^2 Z'$  is unbounded at zero strain rate since the square  $(\tilde{\gamma}_{,z})^2$  of the strain rate gradient is not zero when strain rate  $\tilde{\gamma}$  is zero. In addition,  $\tilde{\gamma}_{,zz}$  is not zero when  $\tilde{\gamma}$  is zero, so the product  $Z\tilde{\gamma}_{,zz}$  is also unbounded at zero strain rate, due to the unboundedness of viscosity  $Z$ . Consequently,  $L_1$  is unbounded at the maxima and minima of oscillations. This is illustrated by the numerical computation of  $L_1$  shown in Fig. 8, where we overlay  $L_1$  on the jet profile for the strain-rate thinning fluid of Fig. 6; similar behavior occurs in the strain-rate thickening solution of Fig. 6. This diagnostic indicates a failure of the asymptotic approximation induced purely by the behavior of the power law form near zero strain rate: The isolated spikes in  $L_1$  invalidate any power law solution of the leading order problem as a leading order, slowly



**Fig. 8** Leading order free surface profile  $\phi_1(z)$  of Fig. 6 with  $l=0.5$  (strain-rate thinning, ----) and correction term  $L_1/\varepsilon^2$  (—), showing spikes at locations where  $\phi_{1,z}=0$ . The spikes in  $L_1/\varepsilon^2$  have amplitudes in the numerical simulation between 95 and  $5.5 \times 10^{16}$

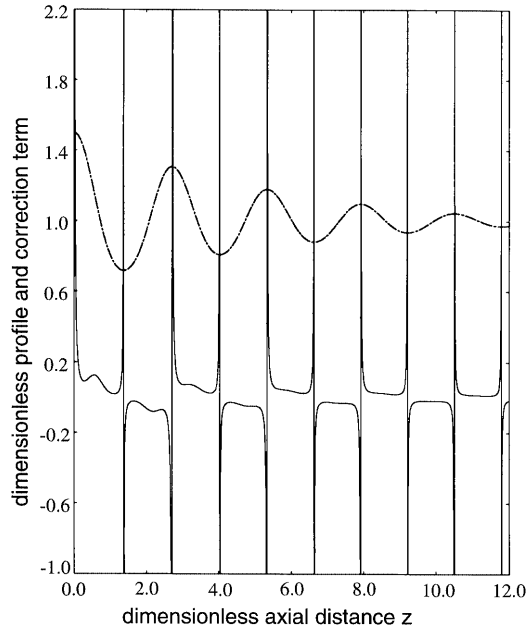
varying approximation to the 3-D behavior by disordering the presumed asymptotic expansion. The spikes falsely indicate the onset of small length scales and breakdown of the asymptotic approximation. The analysis of the 3-D instability of an oscillating jet described by this viscosity form is beyond the scope of this paper, but is irrelevant: physical fluids are observed to establish oscillating jet flows without the onset of small length scales, and hence the power law cannot characterize these fluids in this flow.

#### Cross model

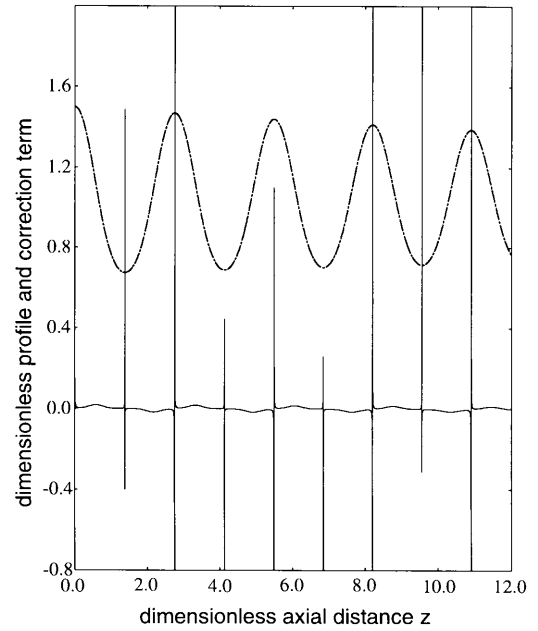
Although the Cross model was constructed to avoid the problems of the power law model at zero strain rate, we again encounter (Fig. 9) artificial large-amplitude spikes in the next order corrections at locations of zero strain rate. This is due to the unbounded viscosity gradient  $\frac{d\eta}{d(\Pi_D)}$  ( $Z'$  in dimensionless form) at zero strain rate in the Cross model (see Eq. 224).

#### Carreau model

The numerical simulation shown in Fig. 10 for an oscillating jet of a strain-rate thinning fluid with viscosity modeled by the Carreau form again indicates large spikes in the correction terms. While the Carreau model yields viscosity and viscosity gradients that are



**Fig. 9** Leading order free surface profile  $\phi_1(z)$  (----) and correction term  $L_1/\epsilon^2$  (—) for an oscillating jet with strain-rate thinning viscosity given by the Cross model:  $W=1$ ,  $Z_0=1.0$ ,  $Z_\infty=0.0171$ ,  $Z_C=276$ , and  $l=0.5$ , showing spikes in  $L_1$  at locations where  $\phi_{1,z}=0$ . The spikes have amplitudes between  $63.4$  and  $9.7 \times 10^{12}$



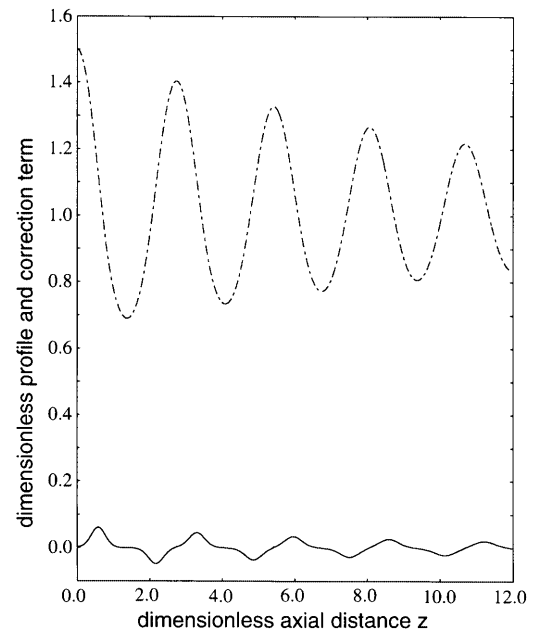
**Fig. 10** Leading order free surface profile  $\phi_1(z)$  (----) and correction term  $L_1/\epsilon^2$  (—) for an oscillating jet with strain-rate thinning viscosity given by the Carreau model:  $W=1$ ,  $Z_0=0.036$ ,  $Z_\infty=0.00541$ ,  $Z_{CY}=1839$ , and  $l=0.5$ , showing spikes in  $L_1/\epsilon^2$  at locations where  $\phi_{1,z}=0$ . The maximum amplitude of the spikes in the numerical simulation is  $40$

finite at zero strain rate (see Eqs. 27) so that  $L_1$  is bounded, the magnitude of  $L_1$  is still large enough to disorder the asymptotic expansion. This is because the dimensionless value of  $Z'$  at zero strain rate in the Carreau form is proportional to the difference between the dimensionless zero strain rate viscosity  $Z_0$  and large strain rate viscosity  $Z_\infty$ , and typically this difference is large. For example,  $Z'(0)=25,900$  in the simulation displayed in Fig. 10 (which models a hypothetical experiment with the material properties  $\rho=1.0 \text{ g cm}^{-3}$ ,  $\sigma=24.2 \text{ dyne cm}^{-1}$ ,  $\eta_0=0.0396 \text{ Pa s}$ ,  $\eta_\infty=0.0057 \text{ Pa s}$ ,  $\lambda=2.09 \text{ s}$ , and  $l=0.5$ , and process conditions of  $r_0=0.05 \text{ cm}$ ,  $z_0=0.5 \text{ cm}$ , and  $v_0=220 \text{ cm s}^{-1}$ ). From Fig. 10 we see that this large value of  $Z'(0)$  produces significant spikes in  $L_1$ .

We comment, without exhibiting the graphs, that the Prandtl-Eyring and Powell-Eyring forms give results similar to but slightly worse than the Carreau form, with large spikes in  $L_1$  at the locations of zero strain rate. This is to be expected, given that they demand a comparable but slightly larger magnitude of viscosity gradient at low strain rates (see Table 1 and Figs. 4 and 5).

### Zhou model

From the analysis above it is clear that we can control the product  $(\dot{\gamma}_z)^2 Z'$  in  $L_1$  at zero strain rate only with a constitutive form in which the viscosity gradient at zero



**Fig. 11** Leading order free surface profile  $\phi_1(z)$  (----) and correction term  $L_1/\epsilon^2$  (—) for an oscillating jet with strain-rate thinning viscosity given by the Zhou model:  $W=1$ ,  $Z_0=0.4545$ ,  $Z_\infty=0.018$ ,  $Z_H=7 \times 10^{-8}$ ,  $\alpha=1.81$ , and  $B=0.05$ . Note the uniformly small magnitude of the correction term, indicating the leading order solution is a valid representation of the 3-D solution

strain rate is zero. The Zhou form (Eq. 34) is constructed expressly to have this property. The simulation in Fig. 11 illustrates that the correction term is bounded everywhere. In fact the spikes are completely absent at the locations of maximum and minimum amplitudes (and zero strain rate); at these locations the correction  $L_1$  is identically zero.

From the analysis and computations of this section we conclude that the power law, Cross, Carreau, Prandtl-Eyring, and Powell-Eyring forms are problematic as characterizations of a strain-rate thinning fluid in flows experiencing locations or times of low strain rates, since they cannot model observed slender oscillating jet behavior: The first order corrections to the leading order solution are either unbounded (for the power law and Cross forms) or large (for the Carreau, Prandtl-Eyring, and Powell-Eyring forms) at the locations where the strain rate is passing through zero; these isolated spikes indicate that if the fluid viscosity was in fact governed by the assumed form, an oscillating jet could not be established, in opposition to physical reality. Our proposed Zhou form, on the contrary, yields uniformly small first corrections, and hence passes the test of being able to model oscillating jet behavior.

## Conclusions

Oscillating jets with an elliptical cross-section provide accurate means for material characterization of other-

wise difficult properties, namely dynamic surface tension and elongational viscosity. Here we have focused on the relevance of oscillating jet models that accompany this experimental technique for the specific case of strain-rate thinning and strain-rate thickening viscous fluids. We have shown analytically and illustrated numerically that standard viscosity forms (the power law, Cross, Carreau, Prandtl-Eyring, and Powell-Eyring forms) for strain-rate thinning inelastic fluids break down when applied to oscillating jet flows in which gravity is a weak effect, precisely because of behavior of these forms near zero strain rate. We then propose a new form that can fit data at least as well as the Cross, Carreau, Prandtl-Eyring, and Powell-Eyring forms, and is in addition consistent with the asymptotic model. These results offer a new model for material characterization experiments on strain-rate thinning fluids.

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