Lecture 1: Introduction

E. J. Hinch

Non-Newtonian fluids occur commonly in our world. These fluids, such as toothpaste, saliva, oils, mud and lava, exhibit a number of behaviors that are different from Newtonian fluids and have a number of additional material properties. In general, these differences arise because the fluid has a microstructure that influences the flow. In section 2, we will present a collection of some of the interesting phenomena arising from flow nonlinearities, the inhibition of stretching, elastic effects and normal stresses. In section 3 we will discuss a variety of devices for measuring material properties, a process known as *rheometry.*

1 Fluid Mechanical Preliminaries

The equations of motion for an incompressible fluid of unit density are (for details and derivation see any text on fluid mechanics, e.g. [1])

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla)\,\mathbf{u} = \nabla \cdot \mathbf{S} + \mathbf{F} \tag{1}$$

$$\nabla \cdot \mathbf{u} = 0 \tag{2}$$

where $\mathbf{u}$ is the velocity, $\mathbf{S}$ is the total stress tensor and $\mathbf{F}$ are the body forces. It is customary to divide the total stress into an isotropic part and a *deviatoric* part as in

$$\mathbf{S} = -p\mathbf{I} + \boldsymbol{\sigma} \tag{3}$$

where $\operatorname{tr}\boldsymbol{\sigma} = 0$. These equations are closed only if we can relate the deviatoric stress to the velocity field (the pressure field satisfies the incompressibility condition). It is common to look for local models where the stress depends only on the local gradients of the flow: $\boldsymbol{\sigma} = \boldsymbol{\sigma}(\mathbf{E})$ where $\mathbf{E}$ is the *rate of strain tensor*

$$\mathbf{E} = \frac{1}{2}\left(\nabla\mathbf{u} + \nabla\mathbf{u}^T\right), \tag{4}$$

the symmetric part of the the velocity gradient tensor.

The trace-free requirement on $\boldsymbol{\sigma}$ and the physical requirement of symmetry $\boldsymbol{\sigma} = \boldsymbol{\sigma}^T$ means that there are only 5 independent components of the deviatoric stress: 3 shear stresses (the off-diagonal elements) and 2 normal stress differences (the diagonal elements constrained to sum to 0). These two normal stress differences are

$$N_1 = \sigma_{xx} - \sigma_{yy} \tag{5}$$

$$N_2 = \sigma_{zz} - \sigma_{yy}. \tag{6}$$

Throughout this series of notes we will frequently refer to two model flow types: *Simple shear* and *Uni-axial extension.* In simple shear the velocity profile is $\mathbf{u} = \dot{\gamma}\,(y, 0, 0)$ where $\dot{\gamma}$ is the *shear rate.* The rate of strain tensor in this case is

$$\mathbf{E} = \begin{pmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

For uni-axial extension, $\mathbf{u} = \dot{\epsilon}\,(x, -y/2, -z/2)$ with rate of strain tensor

$$\mathbf{E} = \dot{\epsilon} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1/2 & 0 \\ 0 & 0 & -1/2 \end{pmatrix},$$

where $\dot{\epsilon}$ is the magnitude of the strain. Note that $\dot{\gamma}$ and $\dot{\epsilon}$ are both scalars, whereas $\mathbf{E}$ is a tensor.

2 Phenomena

2.1 Non-linear Flow

In the simple example of flow down a pipe at low Reynolds numbers, the flow rate in Newtonian fluids increases linearly with the applied pressure drop (see figure 1). Any fluids which deviate from this relation are then non-Newtonian. These fluids can be further classified depending on how this relation changes, relative to the Newtonian example. *Shear thinning* fluids become less viscous with increasing shear rates and so have larger than linear growth with pressure-drop in the flow rate. The microstructures of such materials are smashed up at higher shear. This results in lower viscosities, hence the fluid flows more easily. *Shear thickening* fluids become more viscous with increasing shear rate and hence have less than linear flow rates. Shear thickening behavior is less common and generally arises in fluids that have a highly regular microstructure at rest. When the fluid begins to move, the microstructural components jam against each other, thickening the fluid thus preventing movement. Finally there are *yield* fluids for which there is no flow below a certain critical pressure drop. Some common yield fluids are ketchup, toothpaste, silicate-rich lava and mud. The viscous properties of all of these fluids are strongly dependent on temperature and pressure.

2.2 Inhibition of Stretching

Another phenomenon associated with some non-Newtonian fluids is a dramatic resistance to stretching of fluid elements compared to Newtonian fluids. Typically, the force required to stretch the fluid is ~ 1000 times greater than that required to shear it. Measurements of the *extensional viscosity*, the resistance of the fluid to stretching motions, show large variations in behavior depending on the type of flow (see figure 2).

The high extensional viscosities present in these fluids give rise to a multitude of consequences. Bubbles rising in these fluids form cusps at the downstream end, thus avoiding the large stretching flow out of a rear stagnation point (see figure 3). A similar effect in

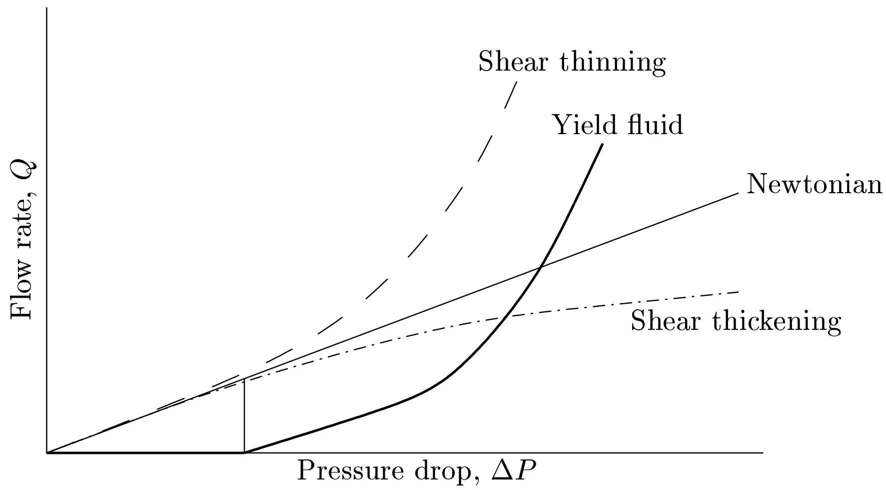

Figure 1: Flow rate as a function of pressure drop for flow down a pipe.

jets inhibits the ejection of spray. The formation of droplets is a highly straining event that occurs at the pinching off of a parcel of fluid. In a fluid containing a few parts per million of high molecular weight polymers, this effect is inhibited by the high extensional viscosity (see figure 4). This lack of spray formation could provide a beneficial effect for fire hoses and in aircraft fuel by preventing potentially explosive mists of droplets from forming.

The inhibition of stretching by high extensional viscosities is also thought to be important in the process of turbulent drag reduction. Addition of very small amounts of high molecular weight polymers to turbulent fluid flows can dramatically reduce the amount of drag in pipe flows. Drag reductions of 50% are possible with polymer concentrations of 10 parts per million (ppm) and as much as an order of magnitude reduction with concentrations of only 500 ppm. This reduction is not well understood and is a much-debated issue in current research. One hypothesis is as follows: drag in turbulent flows is largely due to turbulent bursting events which transport low momentum fluid from near the walls into the interior of the pipe. These bursts are highly straining flows and so are less frequent with the addition of polymers and the attendant increase in extensional viscosity. Such reductions in drag can be critical for oil pipelines (the trans-Alaskan pipeline) and ancient municipal sewer systems. (In Bristol, so great was the drag reduction after a rainfall that a hydrodynamic shock wave was formed in the sewer system and propagated down the network blowing off manhole covers as it passed.)

Conversely, non-Newtonian effects can be detrimental for some industrial processes, for example through the formation of upstream vortices (see section 6 from Lecture 3). Consider flow from a reservoir out a hole: as seen in figure 5, Newtonian fluids flow toward the hole from the entire reservoir while non-Newtonian fluids can form recirculating vortices upstream. These upstream vortices are industrially important in the processing of polymers because fluid that stays in the tank for longer can be significantly degraded (e.g. by a longer exposure to heating), and hence can lead to the production of inconsistent materials. These upstream vortices are caused by high extensional viscosity in the following way: the stretching of fluid elements is proportional to the width of the cone through which fluid flows

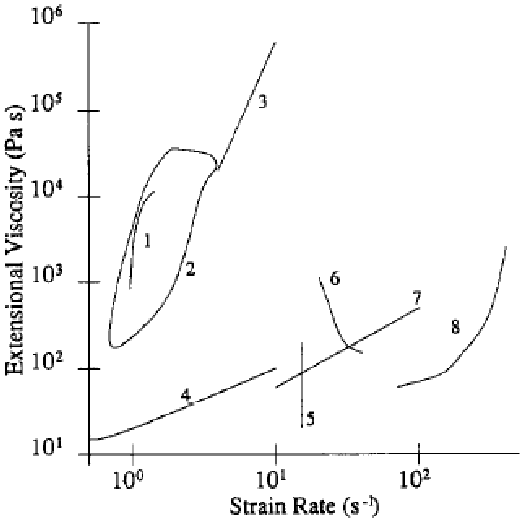

Figure 2: Extensional viscosity measurements for the M1 Boger fluid from (1) open siphon, (2) spinline, (3) contraction flow, (4) opposing jets, (5) falling drop, (6) falling bob, (7) contraction flow, (8) contraction flow [from [2]].

into the hole. For non-Newtonian fluids shear is preferential to stretching and a narrower cone of extensional flow forms at the cost of recirculating vortices ([3]). On the other hand, for Newtonian fluids the cone of fluid flowing out through the hole fills the entire container.

2.3 Elastic Effects

Many non-Newtonian fluids are called visco-elastic because they exhibit a variety of elastic effects in which straining of the fluid can store energy. A dramatic example is shown in figure 6 which shows the cutting of visco-elastic liquid as it is poured. The lower portion of the fluid falls as expected, however the upper portion rebounds upward into the container from which it is being poured. Another interesting effect is the open (tubeless) siphon, in which fluid is drawn up over the wall of the upper container by elastic forces from the descending fluid (see figure 7). Finally in flow out of an orifice, non-Newtonian fluids show an expansion of the stream of fluid known as *die swell* (see figure 8). This expansion is caused by the release of elastic energy stored in the fluid as it is stretched in the outlet tube. This tension causes a vertical rebounds after the fluid leaves the tube and because of incompressibility the stream must expand in the transverse direction.

2.4 Normal Stress Effects

Our final category of non-Newtonian effects contains those caused by stresses normal to shear flow. These effects can be viewed as being due to tension in the streamlines of the flow. For example, there can be dramatic effects on the distribution of particles in shear flows. In simple shear there is aggregation of particles. The tension present in the curved streamlines surrounding two particles produces a net force on the particles that pushes

(a) (b) (c)

Figure 3: Photographs of air bubles in (a) a newtonian fluid (b-c) a non-newtonian fluid (front and side view). Notice the asymmetry.

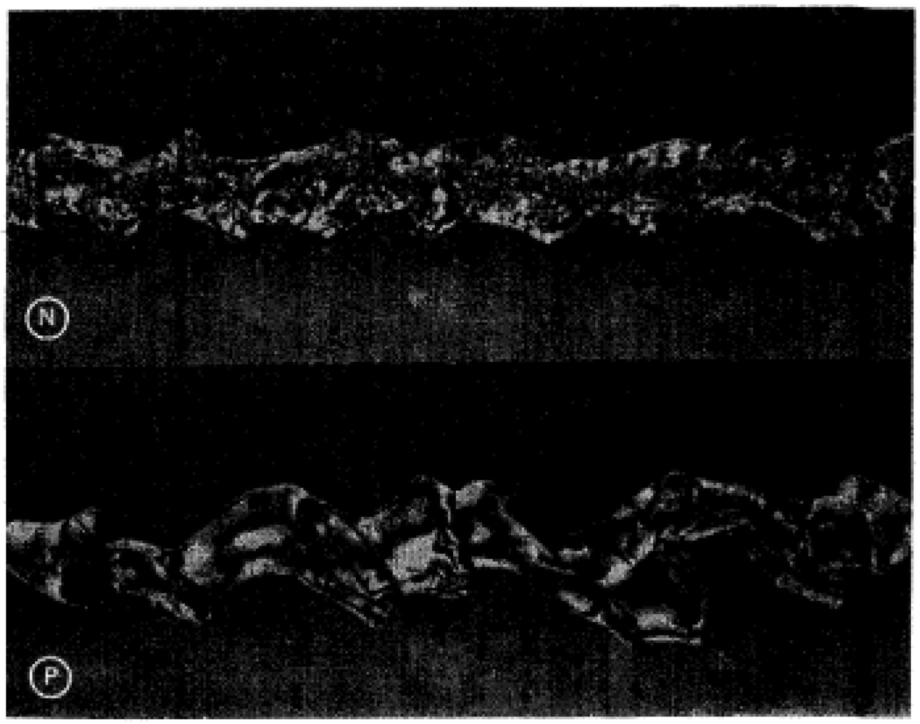

Figure 4: Photograph of high-speed jets for pure water and 200 ppm polyethyleneoxide in water.

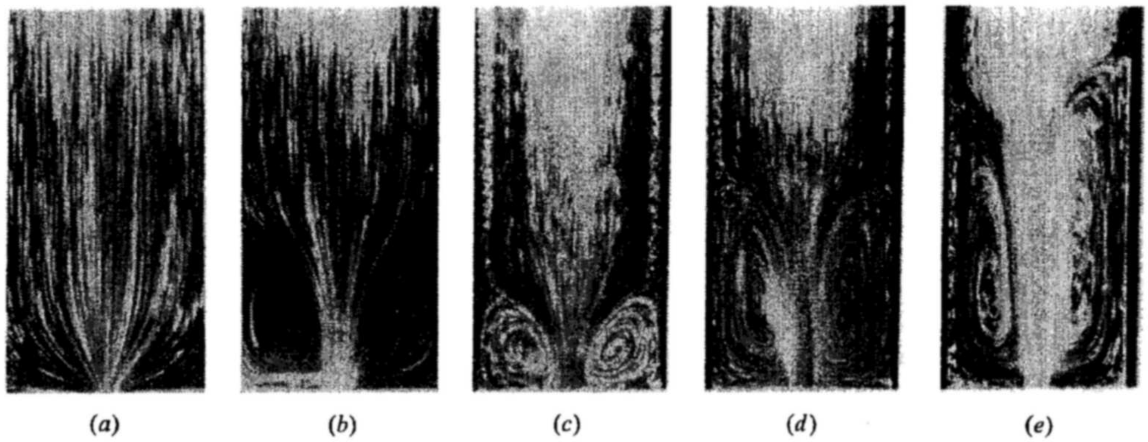

Figure 5: Streamlines of flow out a hole for (a) glycerin, $De = 0$ and (b-e) i for 1.67% aqueous polyacrylamide solution, $De = 0.2$, 1, 3 and 8.

Figure 6: Aluminum soap solution cut in midstream

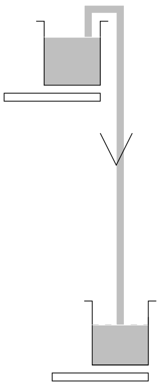

Figure 7: Schematic of the open-siphon effect

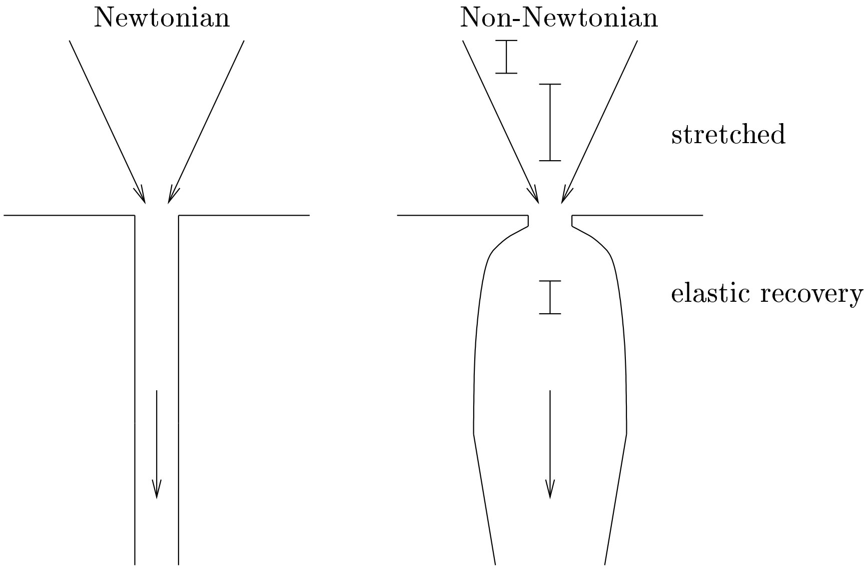

Figure 8: Schematic of the die swell effect for Newtonian and non-Newtonian fluids

them together (figure 9) with a cumulative effect as shown in figure 10. A similar effect is the migration of particles to the center of a pipe. The parabolic velocity profile gives a non-uniform shear that is higher near the walls of the pipe than in the center. This makes the tension in the streamlines greater near the wall and thus applies a net force which causes particles to migrate towards the center as in figure 11.

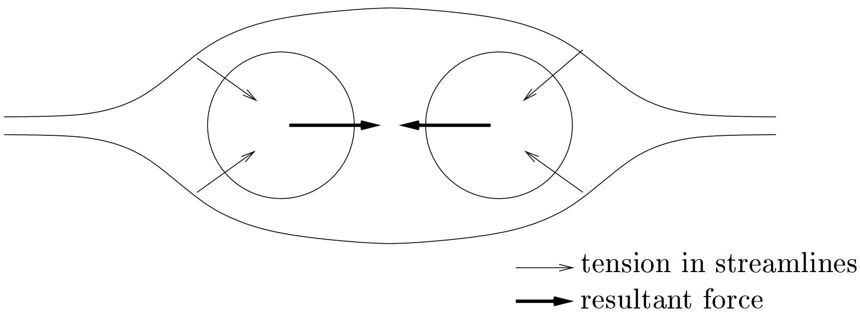

Figure 9: Balance of forces for two particles in a simple shear.

A final example of the effect of normal stresses is that of a spinning rod in a bath of fluid. For low rates of rotation, a Newtonian fluid will have a flat (or slightly depressed) free surface. For comparable rates of rotation in the non-Newtonian fluid, we see an upwards deflection in the free surface, which is higher in the center (see section 4 from Lecture 3). The shear caused by the rotating rod creates tension in the circular streamlines. This "hoop" stress balances the hydrostatic pressure of a column of fluid above it, allowing the fluid to "climb" the rod as in figure 12.

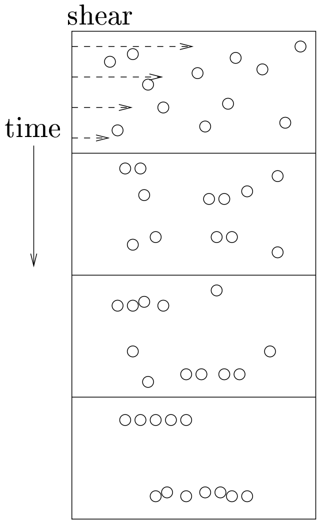

Figure 10: Particle aggregation in sheared polymer solution [after [4]].

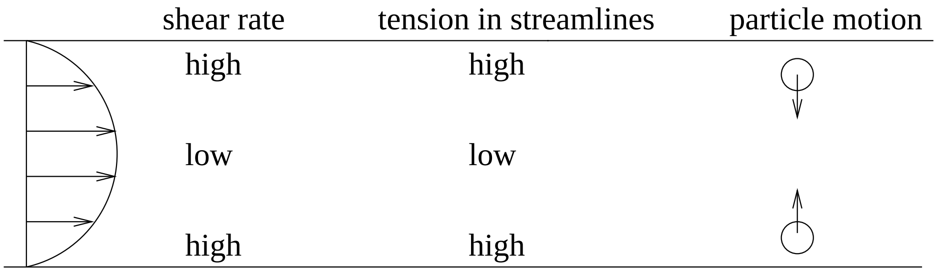

Figure 11: Migration of particles to centerline in a non-Newtonian pipe flow.

3 Rheometry

Rheometry is the study of material properties of fluids including shear viscosities, extensional viscosities and normal stresses as well as the dependence of those properties on temperature and pressure. In this section we discuss the definitions of these properties and the mechanisms used to measure them.

3.1 Simple Shear Devices

There are many ways to generate a shear flow in the laboratory that allow us to measure fluid properties. One of the simplest is shown in figure 13. The fluid lies between two parallel plates with the top plate free to move under an applied force and the bottom plate held fixed. This method works for fluids, such as heavy tars, which are sufficiently viscous so they do not flow out of the sides. The top plate is dragged at constant velocity v across the fluid and feels a force F. The area of the plates is A and their separation is h. The shear rate across the layer is

$$\dot{\gamma} = \frac{v}{h}.$$

Figure 12: A photograph of the rod climbing effect. The device consisted of a rod immersed in the lower, darker fluid. As the rod is rotated, normal stresses cause a fluid column to rise near the rod.

Possible values of this shear rate range from $\dot{\gamma} \approx 10^{-5}\,\mathrm{s}^{-1}$ for fine particles sedimenting, $\dot{\gamma} \approx 10^{1}\,\mathrm{s}^{-1}$ for food being chewed and as high as $\dot{\gamma} \approx 10^{7}\,\mathrm{s}$ in lubrication shear flows. The tangential shear stress is

$$\sigma_{xy} = \frac{F}{A},$$

and the shear viscosity μ is given by the ratio of these two quantities,

$$\mu = \frac{\sigma_{xy}}{\dot{\gamma}} = \frac{Fh}{Av}.$$

Typical values of μ for non-Newtonian fluids are quite large, for example polymer melts have $\mu \approx 10^{3}\,\mathrm{Pa\,s}$ and molten glass has $\mu \approx 10^{12}\,\mathrm{Pa\,s}$ (for water $\mu = 10^{-3}\,\mathrm{Pa\,s}$). Shear-thinning materials often have approximate power law dependence with shear viscosity as a function of shear rate, that is

$$\mu(\dot{\gamma}) = k\dot{\gamma}^{n-1}, \qquad \text{for } n < 1. \tag{7}$$

For molten polymers $n \approx 0.6$, toothpaste has $n \approx 0.3$ and grease has $n \approx 0.1$.

A variety of other devices exist for measuring shear viscosities and these are summarized below (figure 14). The capillary tube rheometer is used for measurements on low viscosity

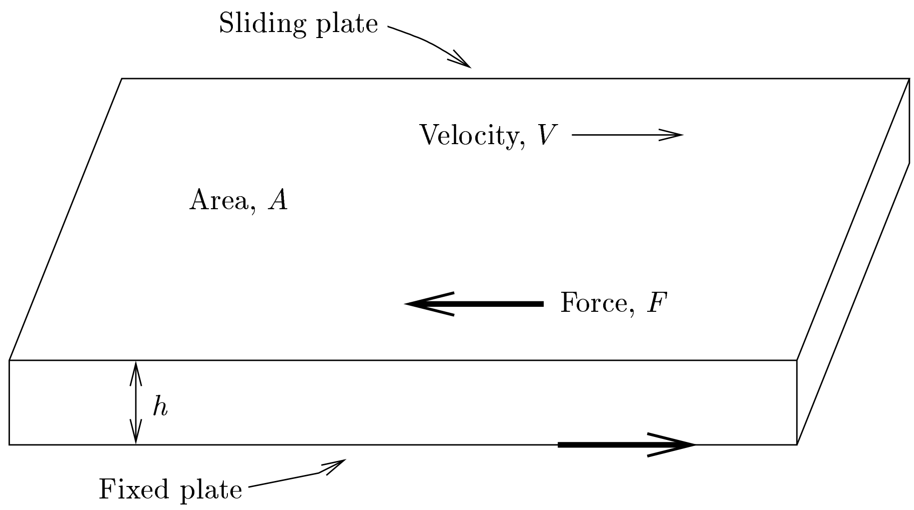

Figure 13: Device for measuring simple shear

liquids with high shear rates. The Couette device is used for flows which have very low Reynolds numbers and so does not suffer from any inertial instability. However, a defect of these two devices is that the shear rate is not uniform throughout the device hence it is not clear what value of $\dot{\gamma}$ is being measured. The cone-and-plate rheometer is designed so that the shear rate is independent of position for small angles $\alpha \approx 2^\circ$ (figure 14). Sample rheometric data are shown in figure 15. The plateau at low shear rates, with power law behavior above a critical value is characteristic of non-Newtonian fluids (see section 3.7).

3.2 Normal Stresses

The normal stresses (the first normal stress due conceptually to the tension in the streamlines) can be measured using the cone-and-plate device described earlier. Tension in the streamlines produces an axial thrust pushing the cone and plate apart with a force which can be measured (see figure 14). With the same device, the second normal stress can be found by measuring the distribution of pressure over the surface of the cone or, if the first normal stress is known, it can be computed from the axial thrust on two rotating parallel plates. A final apparatus for measuring the second normal stress is Tanner's tilted trough, in which non-Newtonian fluid flows down an inclined trough. The free surface is curved due to the influence of the second normal stress and this bowing can be measured with an optical device.

3.3 Oscillatory Rheometry

Rheometers of the parallel plate and cone-and-plate varieties often have the capability to do small amplitude oscillatory shear tests. These tests involve the application of a sinusoidal stress (or strain) to the upper plate or cone of the rheometer. The resulting strain (or stress) can be resolved into components that are in phase with the input (elastic response) and $\frac{\pi}{2}$ out of phase with the input (viscous response). From these data a complex modulus,

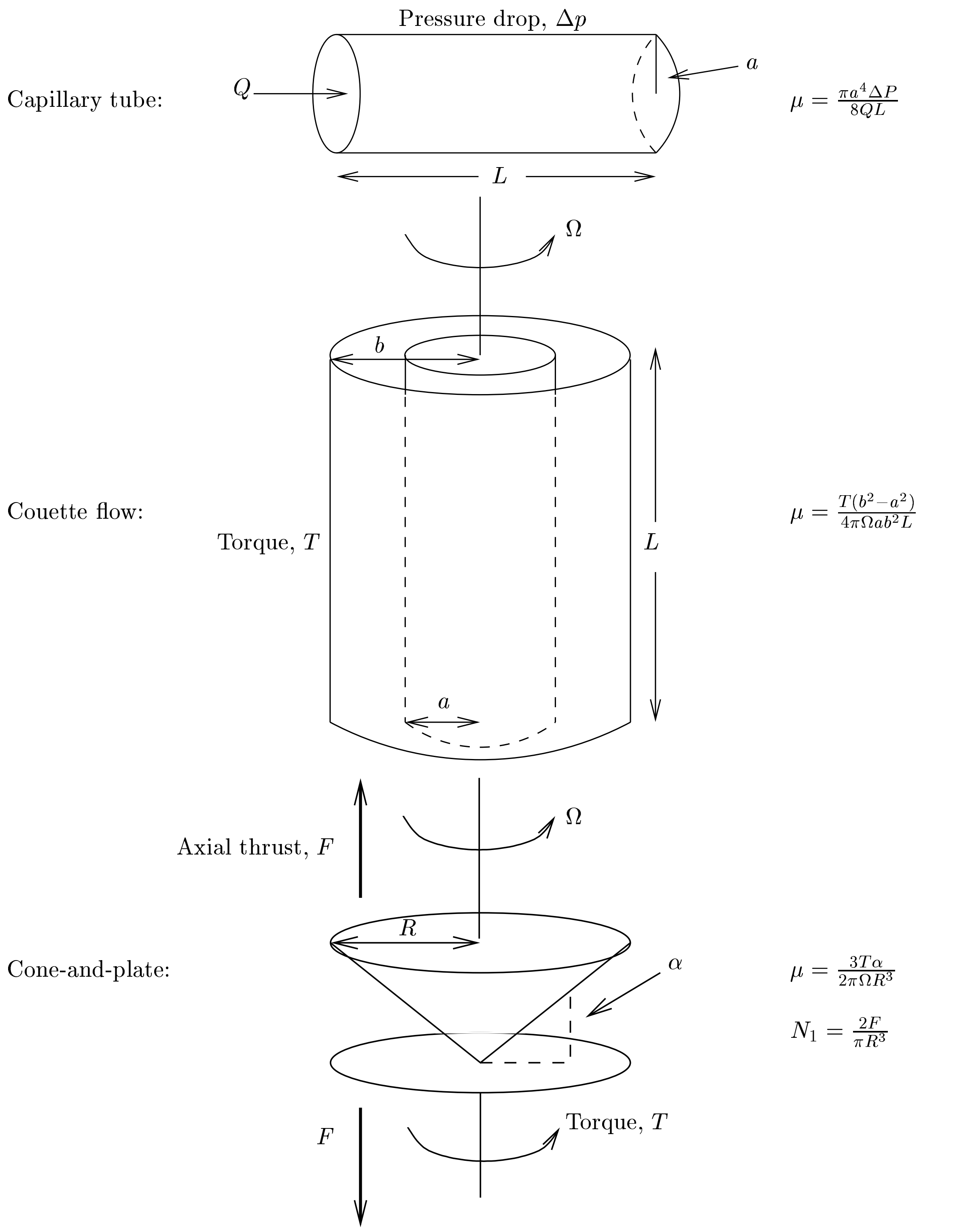

Figure 14: Illustration of devices for measuring shear viscosities. The vertical scale in the cone-and-plate illustration is exaggerated.

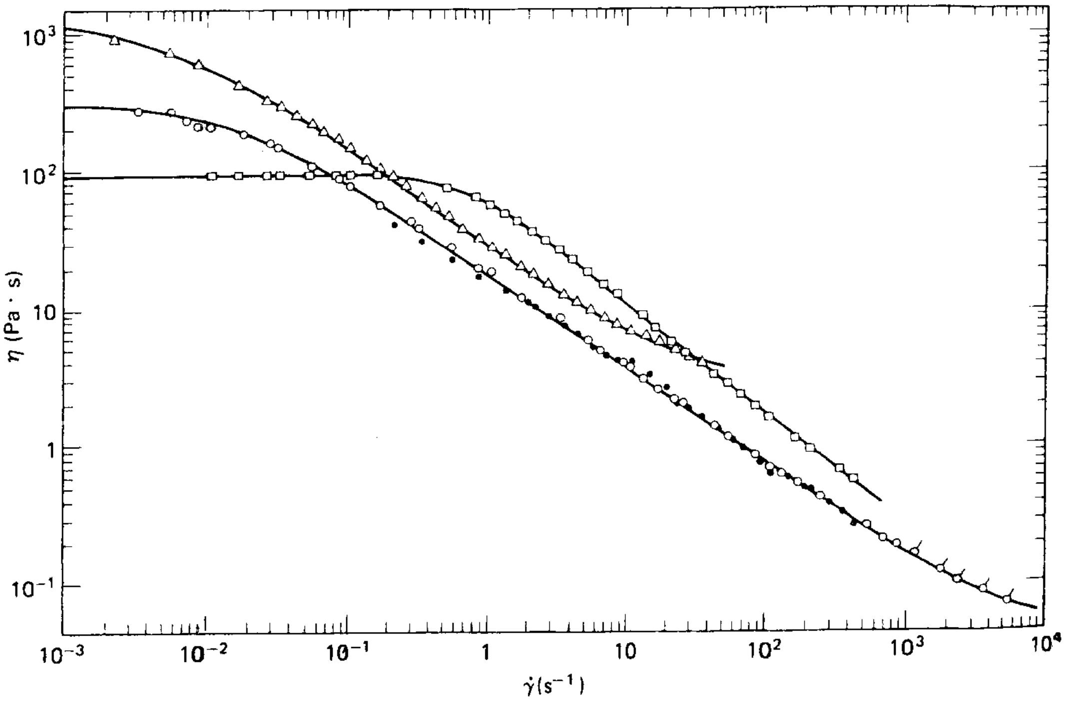

Figure 15: Dependence of viscosity on shear rate for two polymer solutions (∘ and Δ) and an aluminum soap solution (□). All data were taken at 298 K.

G^*, is determined as a function of frequency.

$$G^* = G' + iG'', \tag{8}$$

where G' (storage modulus) and G'' (loss modulus) give information on energy storage and energy dissipation in the flow, respectively. For a perfectly elastic solid, $G'' = 0$ and $G' = G$, the elastic modulus. For a Newtonian fluid, $G' = 0$ and $\mu = \frac{G''}{\omega}$, where ω is the frequency.

3.4 Extensional Viscosity

For the uni-axial extensional flow, we can define the extensional viscosity as,

$$\mu_{\text{ext}} = \frac{\sigma_{xx} - \frac{1}{2}\sigma_{yy} - \frac{1}{2}\sigma_{zz}}{3\dot{\epsilon}}, \tag{9}$$

where σ_{xx}, σ_{yy} and σ_{zz} are the diagonal components of the stress tensor. Unfortunately, in the laboratory this steady straining flow cannot be maintained indefinitely. An approximation to this flow is the spinline experiment (figure 16) where, at every point in the flow, there is one straining direction, in this case the x-direction, and two contracting directions. (for further details see section 7 from Lecture 3 and 1 frpm Lecture 8). Using a similar approximation to that used in section 3.1 we can compute an average stress by dividing the tension T by the area A and an average shear from the velocity gradient $\nabla u \approx (v_2 - v_1)/L$. Then the extensional viscosity is given by

$$\mu_{\text{ext}} \approx \frac{TL}{A(v_2 - v_1)}. \tag{10}$$

Other devices to measure extensional viscosity include the filament stretching rheometer and the Moscow rheometer. The filament stretching rheometer works by placing a fluid between two plates which are pulled apart rapidly (2 m within a second) at a constant strain rate and the applied force on the bottom plate is measured. The Moscow rheometer allows surface tension to squeeze a filament of fluid and measures the rate of thinning. The "Worthington jet" could also be used as a possible method to measure the inhibition of stretching: a solid sphere is dropped into a fluid, as it breaks the surface a cavity forms and the filling of this cavity creates an upwards jet. In non-Newtonian fluids the extensional viscosity retards the motion of the drop and the rebound of the surface [5]. Theory to describe the correlation between the maximum height of this jet and the extensional viscosity has yet to be developed. Other devices to measure extensional viscosity include flow between four rollers or opposed jets, film blowing and Meissner's film on an expanding square grid.

3.5 Temperature, concentration and molecular weight scaling

Material properties depend on a variety of parameters, including the concentration and molecular weights of the polymers and also temperature. Using an appropriate choice of non-dimensional parameters the data may be collapsed to give a power-law dependence for viscosity as a function of shear rate. Figure 17 shows a plot of the non-dimensional reduced viscocity and reduced shear rate, which are defined as

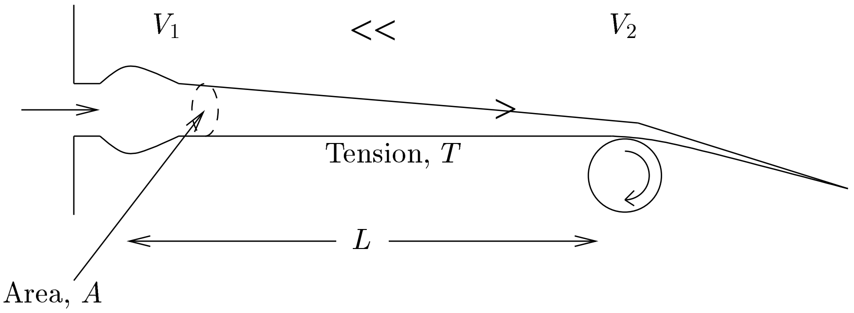

Figure 16: Spinline apparatus for measuring extensional viscosity

$$\mu_r = \mu(\dot{\gamma}, T)\frac{\mu(0, T_*)}{\mu(0, T)}, \tag{11}$$

$$\dot{\gamma}_r = \dot{\gamma}\frac{\mu(0, T)}{\mu(0, T_*)}\frac{T_*\rho_*}{T\rho}, \tag{12}$$

where T_* and ρ_* are a reference temperature and density, respectively. Similarly, figure 18 shows a plot of dimensionless viscosity against dimensionless shear rate for a series of solutions with different concentrations of polymers. Figure 19 shows the power law dependence of viscosity on molecular weight. In the dilute regime, (lower molecular weights) the dependence is linear and in the entangled regime (higher molecular weights) the viscosity is proportional to the molecular weight to the (empirically determined) 3.4 power. The significance of these scalings is that the rheological properties can be determined at a reference condition and then extrapolated to other conditions.

3.6 Cox-Merz rule

The Cox-Merz rule is an empirical rule which states that the dependence of the steady shear viscosity on the shear rate can be estimated from the dynamic viscocity (see section 3.3) as a function of frequency as the two curves are approximately identical (figure 20). This has important practical applications as it is easier to acquire data over a wide range of oscillation frequencies. We force a fluid periodically with frequency ω so that the strain $\gamma(t) = \gamma e^{i\omega t}$ and write the resulting stress as

$$\sigma(t) = G^*(\omega)\gamma(t) = \left(G' + iG''\right)\gamma e^{i\omega t},$$

where G^* is a complex elastic modulus. We can also write

$$\sigma(t) = \mu^*\dot{\gamma}(t) = \left(\mu' + i\mu''\right)i\omega\gamma e^{i\omega t}$$

for a complex viscosity μ^*. The Cox-Merz rule states that $\mu = |\mu^*|$ and $N_1 = 2G'$.

3.7 Non-dimensional Parameters

All materials have a relaxation time τ, the time required to return to its base state after being perturbed, for instance by stretching. This timescale can be seen in figure 3 as the

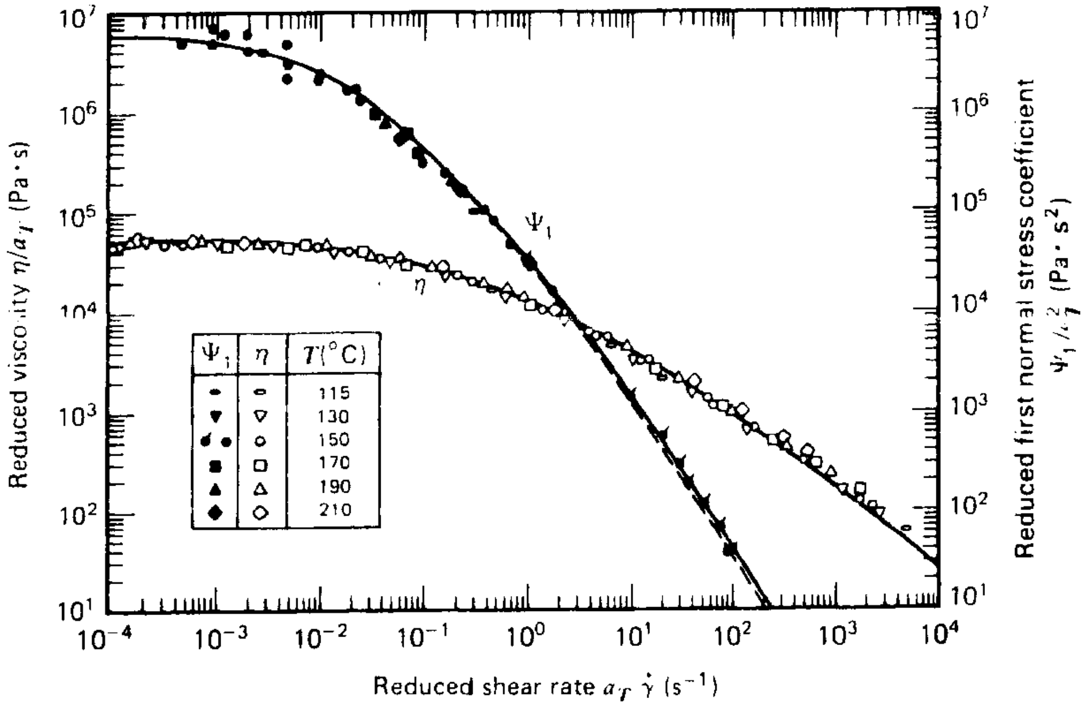

Figure 17: Dimensionless viscosity and first normal stress difference plotted against dimensionless shear rate for a variety of temperatures.

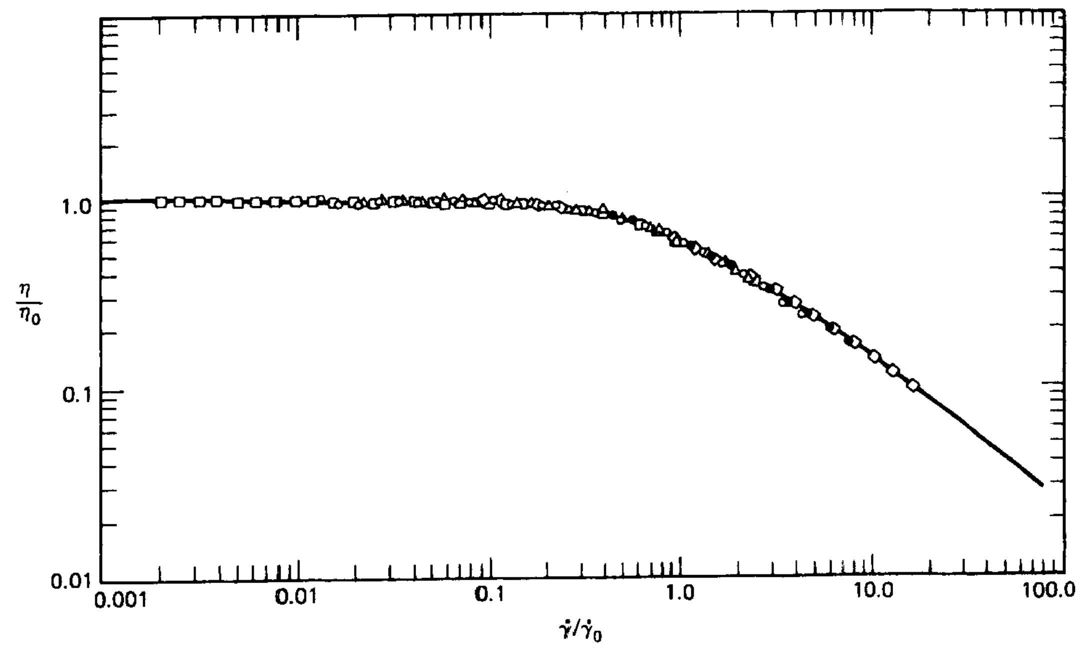

Figure 18: Dimensionless viscosity plotted against dimensionless shear rate for a series of solutions with different solution concentrations.

reciprocal of the shear rate at which the graph of viscosity versus shear rate begins to turn over. In non-Newtonian fluid flow the ratio of the timescales of deformation and relaxation is important. Two important non-dimensional parameters that express this quantity are the Weissenberg number and the Deborah number. The Weissenberg number is a measure of the strength of the shear rate and is defined by

$$Wi = \dot{\gamma}\tau.$$

The Deborah number is the ratio of the characteristic time-scale of the flow to the relaxation time,

$$De = \frac{U\tau}{L}.$$

Note that the Deborah and Weissenberg numbers are often the same (but not always) and either can be used to quantify the importance of relaxation in the fluid. For $De \ll 1$ the material relaxes relatively quickly and it behaves like a viscous fluid. Conversely, when $De \gg 1$ the fluid does not relax on the timescale of the flow and so acts like an elastic solid.

Notes by Neil Burrell and Julia Mullarney

References

[1] G. Batchelor, *An Introduction to fluid dynamics* (Cambridge University Press, Cambridge, 1967).

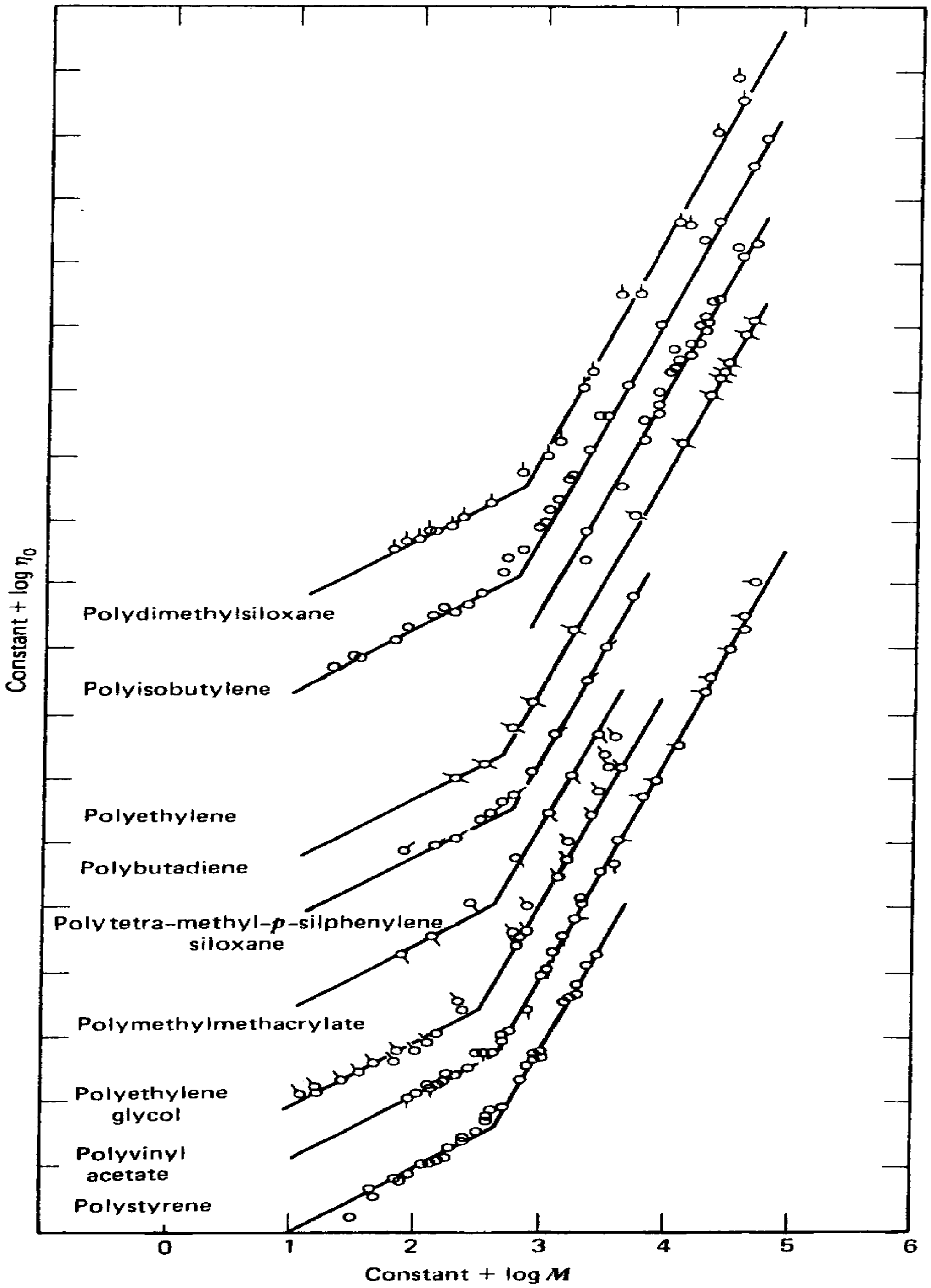

Figure 19: Viscosity versus molecular weight for a variety of polymers.

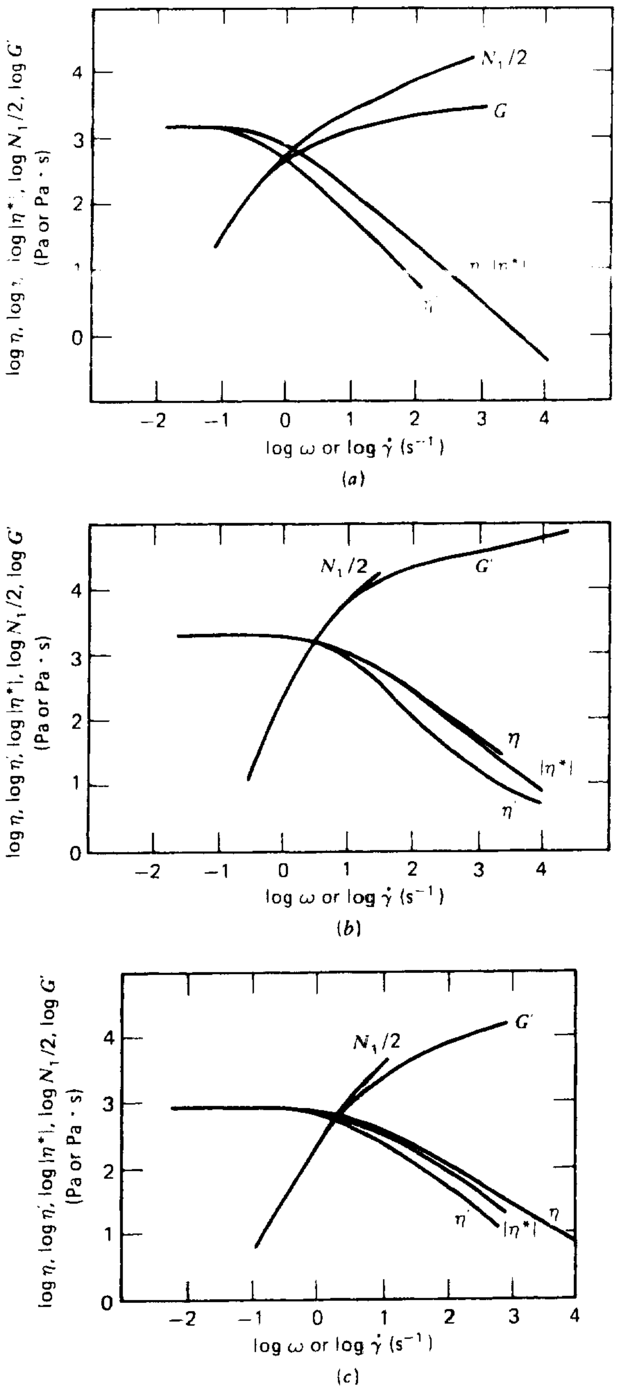

Figure 20: Illustration of the Cox-Merz rule. The curves compare properties under steady shear to their oscillating equivalents.

[2] R. Keiller, “Modelling of the extensional flow of the m1 fluid with the oldroyd equation,” Journal of Non-Newtonian Fluid Mechanics **42**, 49 (1992).

[3] J. R. P. Szabo and E. Hinch, “Start-up of flow of a fene-fluid through a 4:1:4 constriction in a tube,” Journal of Non-Newtonian Fluid Mechanics **72**, 73 (1997).

[4] R. D. J. Michele, R. Pätzold, “Alignment and aggregation effects in suspensions of spheres in non-newtonian media,” Rheological Acta **16**, 317 (1977).

[5] J.-M. Cheny and K. Walters, “Rheological influences on the splashing experiment,” Journal of Non-Newtonian Fluid Mechanics **86**, 185 (1999).