# Fluid Mechanics

Stress Strain Strain rate Shear vs. Extension Apparent Viscosity Oversimplified Models: Maxwell Model Voigt Model Continuity Equation Navier-Stokes Equations Boundary Conditions Volumetric Flow Rate Linear Viscoelasticity

Boltzmann Superposition

Step Strain: Relaxation Modulus Generalized Maxwell Model

Viscosity

Creep/Recovery: Creep Compliance Recoverable Compliance

Steady State Compliance

Terminal Relaxation Time

Oscillatory Shear: Storage Modulus Loss Modulus Phase Angle Loss Tangent

Time-Temperature Superposition

# Molecular Structure Effects

Molecular Models:

Rouse Model (Unentangled) Reptation Model (Entangled)

Viscosity

Recoverable Compliance

Diffusion Coefficient

Terminal Relaxation Time

Terminal Modulus

Plateau Modulus

Entanglement Molecular Weight

Glassy Modulus

Transition Zone

Apparent Viscosity

Polydispersity Effects

Branching Effects

Die Swell

# Nonlinear Viscoelasticity

Stress is an Odd Function of Strain and Strain Rate

Viscosity and Normal Stress are Even Functions of Strain and Strain Rate

Lodge-Meissner Relation

Nonlinear Step Strain Extra Relaxation at Rouse Time Damping Function

Steady Shear

Apparent Viscosity Power Law Model Cross Model Carreau Model Cox-Merz Empiricism First Normal Stress Coefficient Start-Up and Cessation of Steady Shear Nonlinear Creep and Recovery

# Rheometry

Couette Devices:

Gap Loading vs. Surface Loading Controlled Stress vs. Controlled Strain Transducer (and Instrument) Compliance Cone & Plate Parallel Plate Eccentric Rotating Disks Concentric Cylinder Sliding Plates

Poiseuille Devices:

Pressure Driven vs. Rate Driven Capillary Rheometer Wall Shear Stress Wall Shear Rate Bagley End Correction Cogswell Orifice Short-Cut Rabinowitch Correction Slit Rheometer Melt Flow Index Die Swell Extrudate Distortion

# Injection Molding

Injection Molding Cycle Inject and pack mold Extrude next shot once gate solidifies Eject part once part solidifies Injection Molding Economics Only inexpensive if we make many parts Injection Molding Window Poiseuille Flow in Runners and Simple Cavities Calculate injection pressure to fill mold Balance runner systems Calculate clamping force Assumptions: Isothermal Newtonian Hot Runner Systems No runners to regrind More expensive Injection Molding Defects and how to avoid/control them Weld Lines Sink Marks and Voids Short Shots / Uneven Filling Burn Marks Sticking Warping

# Extrusion

Pumping vs. Mixing Pressure Distribution Residence Time Distribution Twin Screw Extrusion Dimensional Analysis Buckingham II Theorem Intuition or Experience Helps Mass Balance

# Uses of Extruders

Injection Molding Blow Molding Pelletizing Heat Transfer Sheet Extrusion Thermoforming Fiber Spinning Pipe Extrusion Film Blowing Coextrusion Barrier properties Profile Extrusion Wire Coating

# Blow Molding

Blow Molding Cycle Parison Extrusion Parison sag Blowing Cooling Ejection Extrusion Blow Molding Economics Less expensive than injection molding Stretch-Blow Molding **Biaxial** orientation **Ring-Neck Blow Molding** Improved thickness control Better precision in neck of bottle Injection-Blow Molding Improved thickness control Fewer surface defects Better precision in neck of bottle Blow Molding Defects and how to minimize them. Advantages of Branched Polymers

# Rotational Molding

The only process we have learned about that does NOT make use of an extruder.

Rotational Molding Cycle High-Speed Rotation to Pack Powder Sintering Cooling (Heat Transfer) Removal

Rotational Molding Economics Cheap way to make small numbers of large parts.

# Thermoset Molding

Gelation: Divergence of Viscosity Growth of Modulus Thermoset Molding Cycle Inject and pack mold Cure part Eject part once part solidifies Thermoset Molding Economics Less capital investment than injection molding No way to recycle waste or final product Compression Molding Transfer Molding Injection Molding **Reaction Injection Molding** Impingement Mixing Solvent Coating Control of Coating Thickness Roll Coating Blade Coating Lubrication Approximation Dip Coating Surface Tension Curtain Coating

#### Stress and Strain







1.Apparent Viscosity of a Monodisperse Polystyrene.

# $\begin{array}{ll} & \text{Oversimplified Models} \\ & \text{MAXWELL MODEL} \\ \text{Stress Relaxation} & \sigma(t) = \sigma_0 \exp(-t/\lambda) \\ & G(t) = G_0 \exp(-t/\lambda) \\ & \text{Creep} & \gamma(t) = \gamma_0(1+t/\lambda) \\ & J(t) = J_s^0(1+t/\lambda) = J_s^0 + t/\eta \\ & J(t) = G'(\omega) = \omega \lambda G''(\omega) = \frac{G_0(\omega \lambda)^2}{1 + (\omega \lambda)^2} \end{array}$

The Maxwell Model is the simplest model of a

VISCOELASTIC LIQUID.

Creep 
$$\gamma(t) = \gamma_{\infty} [1 - \exp(-t/\lambda)]$$
  
 $J(t) = J_{\infty} [1 - \exp(-t/\lambda)]$ 

The Voigt Model is the simplest model of creep for a VISCOELASTIC SOLID.

#### Equations of Fluid Motion

# $\begin{array}{ll} \text{CONTINUITY} \\ \text{Incompressible} & \vec{\nabla} \cdot \vec{v} = 0 \end{array}$

Continuity is a differential equation describing conservation of mass.

#### NAVIER-STOKES

Slow Flows (no inertia,  $R_e < 1$ )  $\rho \frac{\partial \vec{v}}{\partial t} = -\vec{\nabla}P + \rho \vec{g} + \eta \nabla^2 \vec{v}$ 

The Navier-Stokes equations are force balances (per unit volume).

DO NOT MEMORIZE CONTINUITY OR N-S EQUATIONS. IF NEEDED, I WILL GIVE THEM TO YOU.

YOU DO NEED TO KNOW HOW TO USE THEM TO SOLVE

FOR PRESSURE AND VELOCITY DISTRIBUTIONS.

#### **BOUNDARY CONDITIONS**

1. NO SLIP at solid surfaces

2. No infinite velocities

MAXIMUM VELOCITY  
for 
$$v_x = v_x(y)$$
,  $\frac{\partial v_x}{\partial y} = 0$ 

AVERAGE VELOCITY and VOLUMETRIC FLOW RATE  $v_{ave} = \frac{Q}{A} = \frac{1}{A} \int v_x dA$ 

# Linear Viscoelasticity $G(t) \equiv \frac{\sigma(t)}{2}$ Stress Relaxation Modulus BOLTZMANN SUPERPOSITION: Add effects of many step strains to construct ANY linear viscoelastic deformation. $\eta_0 = \int_{0}^{\infty} G(t) dt$ Viscosity $J(t) \equiv \frac{\gamma(t)}{\bar{}}$ Creep Compliance $J_s^0 = \lim_{t \to \infty} \left[ J(t) - \frac{t}{n_s} \right]$ Steady State Compliance $J_s^0 = \frac{1}{n_o^2} \int_0^\infty G(t) t dt$ $R(t)\equiv \frac{\gamma_r(t)}{\sigma}=J(t)-\frac{t}{\sigma}$ **Recoverable Compliance** $J_s^0 = \lim_{t \to \infty} \left[ R(t) \right]$ $\lambda = \eta_0 J_s^0 = \frac{\int_0^\infty G(t) t dt}{\int_{-\infty}^\infty G(t) dt}$ Terminal Relaxation Time

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# Linear Viscoelasticity OSCILLATORY SHEAR RESPONSE OF A LINEAR MONODISPERSE POLYMER



2. Storage and Loss Modulus Master Curves for Polybutadiene at Reference Temperature  $T_0=25\,^oC$ .

Increase 
$$M_w \Rightarrow$$
 Increase  $\lambda$ 

Terminal response is delayed to lower frequency.



3. Storage Modulus of Four Narrow Molecular Weight Distribution Polystyrenes.

Sample	$M_w$
L14	28900
L16	58700
L15	215000
L19	513000



**4.**Storage Modulus Data for Monodisperse Polystyrenes.



**5.**Loss Modulus Data for Monodisperse Polystyrenes.



6. Storage and Loss Moduli for Polystyrene L15 with  $M_w=215000$ .



7. Storage and Loss Moduli for Polystyrene with  $M_w = 315000$  and  $M_w/M_n = 1.8$ .



**8.**Comparison of Monodisperse (L15) and Polydisperse (PS7) Polystyrenes with the Same Viscosity.

#### MOLECULAR THEORIES

#### ROUSE MODEL:

$$D_R \sim \frac{1}{N}$$
  $\lambda_R \cong \frac{R^2}{D_R} \sim N^2$   $G(\lambda_R) = \frac{\rho RT}{M}$   $\eta \cong \lambda_R G(\lambda_R) \sim N$ 

 $G(t) \sim t^{1/2}$  for  $\lambda_N < t < \lambda_R$ 

#### REPTATION MODEL:

Relaxation is simple Rouse motion up to the Rouse relaxation time of an entanglement strand.

$$\lambda_e \sim N_e^2$$
  $G(t) \sim t^{1/2}$  for  $\lambda_N < t < \lambda_e$ 

Plateau Modulus 
$$G_N^0 = \frac{\rho RT}{M_e}$$

D

$$\lambda_d \cong \frac{L^2}{D_R} \sim N^3 \qquad D \cong \frac{R^2}{\lambda_d} \sim \frac{1}{N^2} \qquad \eta \cong \lambda_d G_N^0 \sim N^3$$

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#### Linear Viscoelasticity TIME-TEMPERATURE SUPERPOSITION



Figure 1: (A) Isothermal Storage Modulus  $G'(\omega)$  of a Polystyrene at Six Temperatures. (B) Storage Modulus Master Curve at Reference Temperature  $T_0 = 150$  <sup>o</sup>C.

#### Linear Viscoelasticity OSCILLATORY SHEAR RESPONSE OF A LINEAR MONODISPERSE POLYMER



Figure 2: Storage and Loss Modulus Master Curves for Polybutadiene at Reference Temperature  $T_0 = 25 \ ^{\circ}C$ .

Experimentally,  $G' \sim G'' \sim \omega^u$  with 0.5 < u < 0.8 in the transition zone.

Experimentally,  $\eta_0 \sim M_w^{3.4}$  instead of the reptation prediction of  $\eta_0 \sim M_w^3$ 

Otherwise the molecular theory works fine.

#### Nonlinear Stresses

Shear Stress is an odd function of shear strain and shear rate.

$$\sigma(\gamma) = G\gamma + A_1\gamma^3 + \cdots$$

$$\sigma(\dot{\gamma}) = \eta_0 \dot{\gamma} + A_2 \dot{\gamma}^3 + \cdots$$

Apparent viscosity is thus an even function of shear rate.

$$\eta(\dot{\gamma}) \equiv \frac{\sigma(\dot{\gamma})}{\dot{\gamma}} = \eta_0 + A_2 \dot{\gamma}^2 + \cdots$$

The first normal stress difference is an even function of shear strain and shear rate.

$$N_1(\gamma) = G\gamma^2 + B_1\gamma^4 + \cdots$$

The first term comes from the Lodge-Meissner Relation

$$\frac{N_1}{\sigma} = \gamma$$

$$N_1(\dot{\gamma}) = \Psi_1^0 \dot{\gamma}^2 + B_2 \dot{\gamma}^4 + \cdots$$

First Normal Stress Coefficient is thus an even function of shear rate.

$$\Psi_1 \equiv \frac{N_1(\dot{\gamma})}{\dot{\gamma}^2} = \Psi_1^0 + B_2 \dot{\gamma}^2 + \cdots \cdots$$

#### Nonlinear Step Strain SHORT-TIME RELAXATION PROCESSES



Figure 3: Nonlinear Relaxation Modulus G(t) for a 6% Polystyrene Solution at 30 °C.

#### SEPARABILITY AT LONG TIMES

$$G(t,\gamma) = h(\gamma)G(t,0)$$

$$N_1(t,\gamma) = \gamma^2 h(\gamma) G(t,0)$$

$$h(\gamma) \le 1$$

## Steady Shear

Apparent Viscosity 
$$\eta \equiv$$

 $\frac{\sigma}{\dot{\gamma}}$ 





Figure 4: Shear Rate Dependence of Viscosity and First Normal Stress Coefficient for Low Density Polyethylene.

#### Steady Shear APPARENT VISCOSITY MODELS

Power Law Model  $\eta = \eta_0 |\lambda \dot{\gamma}|^{n-1}$ 

Cross Model  $\eta = \eta_0 \left[ 1 + |\lambda \dot{\gamma}|^{1-n} \right]^{-1}$ 

Carreau Model  $\eta = \eta_0 \left[ 1 + (\lambda \dot{\gamma})^2 \right]^{(n-1)/2}$ 

#### MOLECULAR WEIGHT DEPENDENCES

$$\eta_0 = K M_w^{3.4}$$

$$\lambda = \frac{\eta_0}{G_N^0} \sim M_w^{3.4}$$

$$\Psi_{1,0} = 2\eta_0^2 J_s^0 \sim M_w^{6.8}$$

#### THE COX-MERZ EMPIRICISM

$$\eta(\dot{\gamma}) = |\eta^*(\omega)|$$
  $(\omega = \dot{\gamma})$ 

#### Nonlinear Viscoelasticity START-UP OF STEADY SHEAR



Figure 5: Shear Stress Growth and Normal Stress Growth Coefficients for the Start-Up of Steady Shear of a Polystyrene Solution.

Start-up of nonlinear steady shear shows maxima in shear and normal stress growth functions, indicating extra short-time relaxation processes induced by the large shear rate.

#### Nonlinear Viscoelasticity CESSATION OF STEADY SHEAR



Figure 6: Shear Stress Decay and Normal Stress Decay Coefficients for Cessation of Steady Shear Flow of a Polyisobutylene Solution.

Shear and normal stresses both decay FASTER at larger shear rates, consistent with long relaxation modes being replaced by shorter-time relaxation processes that are activated in steady shear.



Figure 7: Creep Compliance at a Linear Viscoelastic Stress  $\sigma_1$  and two Nonlinear Stresses with  $\sigma_3 > \sigma_2 > \sigma_1$ .

As stress increases, the viscosity drops and the recoverable strain drops, consistent with large stresses inducing additional dissipation mechanisms.



#### NONLINEAR RECOVERY

Figure 8: Recoverable Compliance after Creep at Three Stress Levels (Increasing Creep Stress from Top to Bottom).

#### Nonlinear Viscoelasticity RECOIL DURING START-UP OF SHEAR



Figure 9: Recoil Part-Way Through Start-Up.



Figure 10: Ultimate Recoil During Start-Up Compared with the Shear and Normal Stress Growth Functions for LDPE.

Recoil during start-up of nonlinear steady shear shows a strong maximum because there is a short-time relaxation process activated by the strong shear.

#### Rheometry ROTATIONAL AND SLIDING SURFACE RHEOMETERS GAP LOADING vs. SURFACE LOADING

Compare rheometer gap h to shear wavelength  $\lambda_s = \frac{2\pi}{\omega\sqrt{\rho/G_d}\cos(\delta/2)}$ 

 $\frac{h}{\lambda} \gg 1$ 

Gap Loading Limit:  $\frac{h}{\lambda_1} \ll 1$ 

Surface Loading Limit:

For liquids of high viscosity, the shear wavelength is large and thus we are always in the gap loading limit for polymer melts and concentrated solutions.

TWO CLASSES OF GAP LOADING INSTRUMENTS:

- 1. Impose Strain and Measure Stress
- 2. Impose Stress and Measure Strain

#### INSTRUMENT AND TRANSDUCER COMPLIANCES

$$G'_{a} = \frac{\eta\left(\frac{c\eta}{K} + \lambda\right)\omega^{2}}{\left(\frac{c\eta}{K} + \lambda\right)^{2}\omega^{2} + 1}$$

$$G_a^{"} = \frac{\eta\omega}{\left(\frac{c\eta}{K} + \lambda\right)^2 \omega^2 + 1}$$

For a known instrument/transducer compliance, one may calculate the true moduli of the material from the apparent values.

$$G' + iG'' = \frac{G'_a + iG''_a}{1 - \frac{G'_a}{k} - \frac{iG''_a}{k}}$$

#### Rheometry ROTATIONAL AND SLIDING SURFACE RHEOMETERS

## GEOMETRIES OF GAP LOADING INSTRUMENTS:

1. Cone and Plate



Figure 11: The Cone and Plate Rheometer.

2. Parallel Disks



Figure 12: The Parallel Disk Rheometer.

#### Rheometry CAPILLARY RHEOMETER



Figure 1: The Capillary Rheometer.

Wall Shear Stress  $\sigma_w = \frac{R}{2} \left( -\frac{dP}{dz} \right)$ 

 $\dot{\gamma}_A = \frac{4Q}{\pi B^3}$ 

Apparent Wall Shear Rate



Figure 2: Pressure Distribution in Both the Reservoir and the Capillary.

Bagley correction finds dP/dz in capillary by measuring the end effects through experiments using dies of different length.

$$\sigma_w = \frac{P_d}{2(L/R+e)}$$





Figure 3: Bagley End Correction for Capillary Flow.

#### Rheometry CAPILLARY RHEOMETER

Alternatively, we can use the Cogswell Orifice Short-Cut

$$\sigma_w = \frac{(P_d^L - P_d^0)R}{2L}$$

#### RABINOWITCH CORRECTION

Finally, we plot  $\log \dot{\gamma}_A$  vs.  $\log \sigma_w$  to perform the Rabinowitch correction which calculates the true shear rate at the wall for a general (non-Newtonian) liquid.

$$\dot{\gamma}_w = \left(\frac{3+b}{4}\right)\dot{\gamma}_A$$

$$b \equiv \frac{d(\log \dot{\gamma}_A)}{d(\log \sigma_w)}$$

QUESTION: What happens if the slope of  $\log \dot{\gamma}_A$  vs.  $\log \sigma_w$  is unity for all shear rates? What does this special case correspond to?

$$b \equiv \frac{d(\log \dot{\gamma}_A)}{d(\log \sigma_w)} = 1$$

$$\dot{\gamma}_w = \left(\frac{3+b}{4}\right)\dot{\gamma}_A = \dot{\gamma}_A$$

This case corresponds to a Newtonian liquid, with  $\sigma_w = \eta \dot{\gamma}_w$ . QUESTION: What happens with a shear thinning polymer melt?

$$b \equiv \frac{d(\log \dot{\gamma}_A)}{d(\log \sigma_w)} > 1$$

$$\dot{\gamma}_w = \left(\frac{3+b}{4}\right)\dot{\gamma}_A > \dot{\gamma}_A$$

#### Rheometry SLIT RHEOMETER

The true pressure drop in the slit is measured directly using flush-mounted pressure transducers.



Figure 4: The Slit Rheometer. L > W >> h.

Wall Shear Stress 
$$\sigma_w = \frac{-\Delta P}{L} \frac{h}{2}$$
Wall Shear Rate  $\dot{\gamma}_w = \left(\frac{6Q}{h^2 w}\right) \left(\frac{2+\beta}{3}\right)$ 
$$\beta = \frac{d \left[\log(6Q/h^2 w)\right]}{d \left[\log(\sigma_w)\right]}$$

Apparent Viscosity  $\eta = \frac{\sigma_w}{\dot{\gamma}_w} = \frac{-\Delta P}{L} \frac{h^3 w}{4Q(2+\beta)}$ 



Figure 5: Extrudate Swell after Exiting the Die Diminishes as the Die is Made Longer because the Memory of the Flow Contraction at the Entrance is Reduced.

With a specific polymer and die, die swell increases with increasing shear stress.

Die swell increases as the die is shortened.

Die swell increases as the molecular weight increases.

Die swell increases as the molecular weight distribution is broadened, as it is particularly sensitive to the high molecular weight tail of the distribution.

#### Rheometry SLIT AND CAPILLARY RHEOMETERS EXTRUDATE DISTORTION



Figure 6: Wall Shear Stress vs. Wall Shear Rate for HDPE Showing Flow Instabilities and Wall Slip.

Flow instabilities occur in all rheometers at sufficiently high stress levels.

In cone and plate and parallel disk rotational Couette rheometers, the shear stress required for the onset of flow instabilities is considerably lower than for the Poiseuille flow rheometers.

#### Molecular Structure Effects POLYDISPERSITY



Figure 21: Apparent Viscosity in Steady Shear for Polystyrene. Filled symbols have  $M_w = 260000$  with  $M_w/M_n = 2.4$ . Open symbols have  $M_w = 160000$  with  $M_w/M_n < 1.1$ .

Zero shear viscosity is simply a function of weight-average molecular weight.

$$\eta_0 = \begin{cases} K_1 M_w & \text{for } M_w < M_c \text{ (unentangled)} \\ K_2 M_w^{3.4} & \text{for } M_w > M_c \text{ (entangled)} \end{cases}$$

Steady state compliance, and other measures of elasticity (such as first normal stress difference and die swell) are strong functions of polydispersity.

$$J_s^0 \sim \left(\frac{M_z}{M_w}\right)^a$$
 with  $2 < a < 3.7$ 



Figure 22: Apparent Viscosity of Randomly Branched Polymers Compared to Linear Polymers.

Monodisperse entangled branched polymers have a stronger dependence of viscosity on molecular weight than linear polymers.

$$\eta_0 \sim \exp\left(\frac{\nu M_b}{M_e}\right)$$

Monodisperse entangled branched polymers have steady state compliance increasing with molecular weight.

$$J_s^0 = \frac{0.6M_b}{cRT}$$

 $M_b$  is the molecular weight of the star arm.

Randomly branched polymers have effects of **both** branching and polydispersity.

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# Extrusion

Extruder Characteristic:

$$Q = \alpha N - \frac{\beta}{\mu} \Delta P$$

Die Characteristic:

$$Q = \frac{K}{\mu} \Delta P$$

together, they determine the Operating Point

Pumping vs. Mixing: Compression Ratio and Flow Restrictions

Pressure Distribution

Residence Time Distribution

Twin Screw Extrusion

Dimensional Analysis Buckingham PI Theorem Intuition or Experience Helps

Mass Balance

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