Non-Newtonian Fluid Mechanics

based on a course given by

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Contents

1 Introduction to Non-Newtonian Fluids

Lecture 1
08/10/20

Newtonian fluids are typically characterised by 2 material properties: viscosity and density. We may also refer to Newtonian fluids as 'simple fluids'.

Non-Newtonian fluids may have many other material properties, for example intrinisic time, length and stress scales, and an intrinsic orientation. They often exhibit a mix of fluid and solid behaviour. We may also refer to non-Newtonian fluids as 'complex fluids'.

There are many examples of complex fluids readily apparent in our everyday lives, for example sand (wet or dry) and mud; lava and glass, both of which experience phase changes; ketchup; foam; paint; emulsions such as milk; liquid crystals used in screens; blood on very small scales.

The goal of this course is to cover three main areas.

- Phenomenology of non-Newtonian fluids – how do they behave?

- Mathematical modelling – how do we quantify the behaviour?

- Predictions (and limits) of models

2 Summary of Newtonian Fluid Mechanics

2.1 Continuum approximation

We describe fluids in terms of two main fields: *density* $\rho(\boldsymbol{x}, t)$ and *velocity* $\boldsymbol{u}(\boldsymbol{x}, t)$. We use the continuum approximation whereby the fluid is assumed to be a continuum rather than made up of discrete fluid particles. Under this assumption, the macroscopic properties of density and velocity are well-defined as 'averages' of infinitesimal volume elements.

The velocity field is *Eulerian*, meaning it is measured at a specific point in space and time, as opposed to following a material element (Lagrangian).

2.2 Conservation of mass

Conservation of mass can be expressed in the classical form of a conservation equation as:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot [\rho \boldsymbol{u}] = 0$$

Expanding the flux term, this may be expressed in a form which relates the rate of change of density of a fluid element with the divergence of the flow:

$$\frac{\mathrm{D}\rho}{\mathrm{D}t} = -\rho \nabla \cdot \boldsymbol{u}$$

In this course we will assume that all fluids are incompressible, which is expressed mathematically as:

$$\frac{\mathrm{D}\rho}{\mathrm{D}t} = 0 \iff \nabla \cdot \boldsymbol{u} = 0$$

2.3 Mechanical equilibrium

Newton's second law, i.e. conservation of momentum is expressed for a fluid using the *Cauchy momentum equation*:

$$\rho \frac{\mathrm{D}\boldsymbol{u}}{\mathrm{D}t} = \rho \left[\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} \right] = \nabla \cdot \sigma + \boldsymbol{F}$$

where $\nabla \cdot \sigma$ are the surface forces acting on the fluid and $\boldsymbol{F}$ are body forces which we will assume to be negligible unless otherwise stated.
The Cauchy momentum equation is valid for all continuum fluids. To close the equation, we require an expression for σ.

Definition. The *stress tensor* σ is a symmetric second-rank tensor.
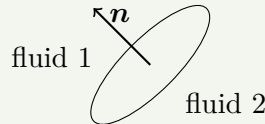
Physically, $\sigma_{ij} n_j$ is the ith component of the force per unit area from the motion of fluid 1 on fluid 2, where $\boldsymbol{n}$ is the surface normal pointing into fluid 1.

2.4 Constitutive modelling

The stress tensor is specified in terms of the deformation via *constitutive modelling*.

Example. Newton's experiment
Consider two parallel plates of area A separated a distance h by a fluid. We consider the force F required on the top plate to induce motion at speed U. Note that there is no force perpendicular to the plates if the fluid is Newtonian. This is not necessarily true for complex fluids.
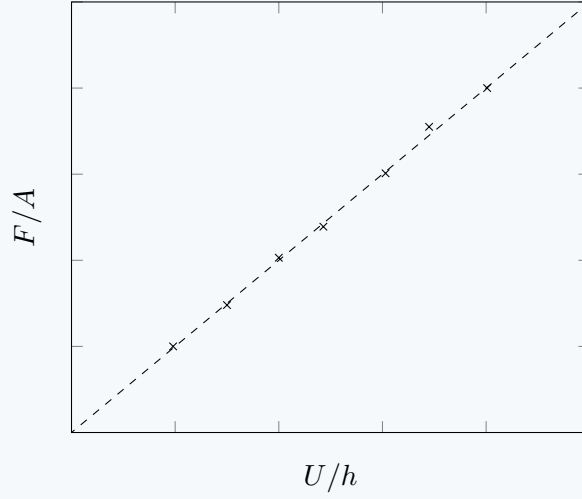
From experiment, we find $\sigma = F/A \propto U/h$. Note that U/h has dimensions of time^{-1}. We define the *shear rate* $\dot{\gamma} = U/h$ and the *viscosity* η via $\sigma = \eta\dot{\gamma}$. Viscosity is a constant material property, for example in water $\eta = 10^{-3}\,\mathrm{Pa}\cdot\mathrm{s}$.

We can now generalise for all Newtonian flows. We start by separating out an isotropic component of the shear tensor:

$$\sigma_{ij} = -p\delta_{ij} + \tau_{ij}$$

where p is the *dynamic pressure* and τ is the *deviatoric stress*. The deviatoric stress may be a function of $u_i, \dfrac{\partial u_i}{\partial x_j}, \dots$; local or non-local in time or space; or a function of other material properties and parameters. For Newtonian fluids, we make five assumptions.

1. Galilean invariance: the deviatoric stress cannot depend on u_i

2. Instaneous response: there is no dependence on the history of deformation

3. Locality: no dependence on second or higher spatial derivatives

4. Linearity: τ_{ij} is linearly related to $\dfrac{\partial u_m}{\partial x_n}$

5. Isotropy: the relationship is independent of reference frame, i.e. isotropic

We can satisfy 1, 2, 3, and 4 by writing

$$\tau_{ij} = A_{ijkl}\frac{\partial u_k}{\partial x_l}$$

where A_{ijkl} is a fourth rank tensor. Using the form of the most general isotropic fourth rank tensor we may enforce isotropy.

$$A_{ijkl} = A\delta_{ij}\delta_{kl} + B\delta_{ik}\delta_{jl} + C\delta_{il}\delta_{jk}$$

Since σ is symmetric, τ is symmetric, therefore $A_{ijkl} = A_{jikl}$. This requires $B = C \equiv \eta$. Thus

$$\tau_{ij} = A\delta_{ij}\frac{\partial u_k}{\partial x_k} + \eta\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)$$

$$= \eta\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)$$

$$= 2\eta e_{ij}$$

since $\frac{\partial u_k}{\partial x_k} = \nabla \cdot \boldsymbol{u} = 0$. Note $e = \frac{1}{2}\left(\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T\right)$ is the *rate of strain* tensor. This is the *Newtonian constitutive relationship*.

> **Definition.** To exclude the factors of 2 we define the *shear rate* $\dot{\gamma}_{ij} = 2e_{ij}$ so that $\tau_{ij} = \eta \dot{\gamma}_{ij}$.

Combining the Cauchy momentum equation and Newtonian constitutive relationship yields the *Navier-Stokes equations*

$$\rho \frac{\mathrm{D}\boldsymbol{u}}{\mathrm{D}t} = -\nabla p + \eta \nabla^2 \boldsymbol{u}$$

Consider a general body with intrinsic length scale L, velocity length scale U and unsteady motion frequency scale ω. Two dimensionless numbers are used to quantify the importance of inertia:

$$\mathrm{Re} = \frac{\rho U L}{\eta}, \qquad \mathrm{Re}_\omega = \frac{\rho \omega L^2}{\eta}$$

We will assume throughout this course that $\mathrm{Re} \ll 1$ and $\mathrm{Re}_\omega \ll 1$ unless otherwise stated. In this no-inertia limit, the Navier-Stokes equations simplify to the *Stokes equations*. For a general fluid these are:

$$\nabla \cdot \sigma = 0$$

Using the Newtonian constitutive relationship, the Stokes equations are:

$$\boldsymbol{0} = -\nabla p + \eta \nabla^2 \boldsymbol{u}$$

Example. Newton's experiment revisited
We will apply the above theory to Newton's experiment and show the same results are obtained.
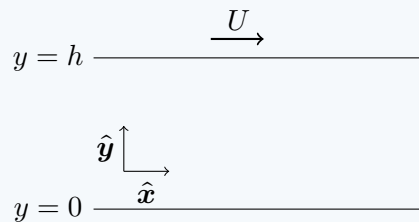
Assume the flow is unidirectional: $\boldsymbol{u} = u(y)\hat{\boldsymbol{x}}$. The Stokes equations become

$$\begin{cases} \dfrac{\partial p}{\partial x} = \eta \dfrac{\partial^2 u}{\partial y^2} & = \text{const.} \\ \dfrac{\partial p}{\partial y} = 0 & \implies p = p(x) \end{cases}$$

Assuming there is no net pressure drop, $p = p(x) \equiv 0$. Applying the boundary conditions we find

$$u(y) = Uy/h \implies \dot{\gamma} = U/h$$

as before. This is a *shear* or *Couette* flow.

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3 Phenomenology

There are four distinguishing properties of non-Newtonian fluids which differ to Newtonian fluids.

3.1 Shear-dependent viscosity

Newtonian fluids have a constant viscosity η which is a constant of proportionality between shear stress σ and shear rate $\dot{\gamma}$. For a complex fluid, η is *not* constant. We re-define viscosity as an implicit function of shear rate.

> **Definition.** Viscosity is defined via $\eta \equiv \frac{\sigma}{\dot{\gamma}} = \eta(\dot{\gamma})$.
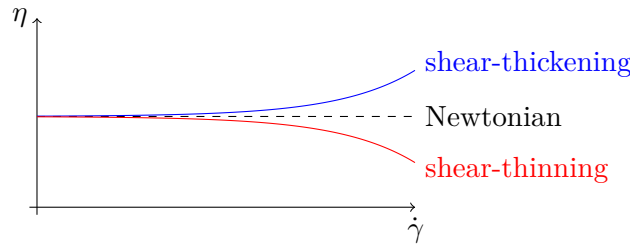
We can broadly categorise complex fluids into two types: *shear thinning* and *shear thickening* fluids.

Examples of shear-thinning fluids are polymer suspensions; paint; blood. These complex fluids have $\frac{\partial \eta}{\partial \dot{\gamma}} < 0$.

Examples of shear-thickening fluids are: cornstarch in water; suspesions of colloidal particles. These complex fluids have $\frac{\partial \eta}{\partial \dot{\gamma}} > 0$.

> **Definition.** The zero-shear rate viscosity is $\eta_0 = \lim_{\dot{\gamma} \to 0} \eta$.

Note that some complex fluids have approximately constant viscosity (Boger fluids).

3.2 Fluid memory

Consider a step-shear flow, that is, Newton's experiment where the top place impulsively starts motion at $t = 0^+$. The response of a Newtonian fluid is on an inertial timescale $\tau \sim h^2/\nu$ which is almost instantaneous, whilst a non-Newtonian fluid takes time to respond to adjust to the change in deformation: the jump occurs over some *relaxation timescale λ*.
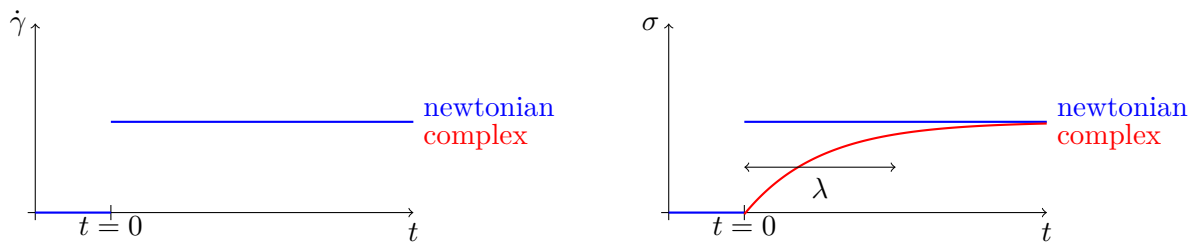
The relationship between stress and deformation is *history dependent*. Impulsively removing the applied shear (i.e. the plate impulsively comes to rest) results in *stress relaxation* with $\sigma \sim e^{-t/\lambda}$.
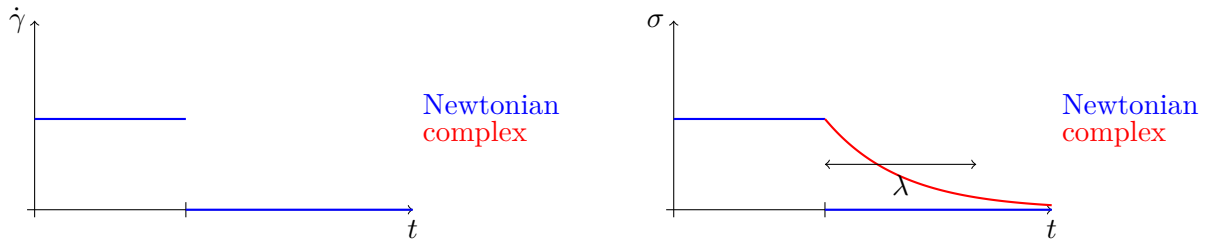
The reverse is also true. Imposing a stress σ and measuring the deformation $\dot{\gamma}$, complex fluids in general wil have a history-dependent response. This is called *strain retardation* and λ' is the *retardation timescale.*

3.3 Normal stress differences

In Newton's experiment, a Newtonian fluid exerts no normal force F_N on the moving plate, since linearity and reversibility ($U \to -U$) impies $F_N = -F_N \equiv 0$. We previously calculated

$$\boldsymbol{u} = \frac{Uy}{h}\widehat{\boldsymbol{x}}, \qquad \dot{\gamma} = \frac{U}{h}$$

Thus in the Newtonian case the stress tensor has the form

$$\sigma = \begin{pmatrix} -p_0 & \eta\dot{\gamma} & 0 \\ \eta\dot{\gamma} & -p_0 & 0 \\ 0 & 0 & -p_0 \end{pmatrix}$$

where p_0 is the external pressure. Note that the normal stresses are equal and constant.
In the non-Newtonian case we have

$$\sigma = \begin{pmatrix} \sigma_{xx} & \eta)(\dot{\gamma})\dot{\gamma} & 0 \\ \eta(\dot{\gamma})\dot{\gamma} & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$$

where in general normal stresses are not equal and not constant. Normal stresses include the external pressure, so the relevant quantity is the difference between normal stresses.

> **Definition.** The first and second *normal stress differences* are
>
> $$N_1 = \sigma_{xx} - \sigma_{yy}, \qquad N_2 = \sigma_{yy} - \sigma_{zz}$$

Note the following.

- For a Newtonian fluid, $N_1 = N_2 = 0$

- Polymeric fluids (for example) have $N_1 > 0, N_2 < 0, |N_2/N_1| \sim 0.1$

- N_1 and N_2 are defined only for steady shear flow

- N_1 and N_2 will, in general, depend on $\dot{\gamma}$

- Boger fluids have constant viscosity but they have non-zero N_1 and N_2

Reversibility implies N_1 and N_2 have to be *even* functions of $\dot\gamma$. In the limit $\dot\gamma \to 0$, i.e. the Newtonian limit, we should have $N_1 \to 0, N_2 \to 0$. Thus the Taylor expansion of N_1, N_2 near $\dot\gamma = 0$ is

$$N_{1,2} = A_{1,2}\dot\gamma^2 + B_{1,2}\dot\gamma^4 + ...$$

Definition. The *normal stress coefficients* are

$$\Psi_1 = \frac{N_1}{\dot\gamma^2}, \qquad \Psi_2 = \frac{N_2}{\dot\gamma^2}$$

The physical consequence of having normal stress differences is the introduction of elastic tension along flow streamlines.

Suppose $\sigma_{xx} = -p$. Thus compression $p > 0 \implies \sigma_{xx} < 0$ and $N_1 > 0 \implies \sigma_{xx} > 0$ which can be thought of as 'negative pressure' which acts as tension. An intuitive example is stretching of polymer molecules. This has many consequences on experiments and flow behaviour.

Example. Two examples of the consequences of normal stress differences are as follows.

1. Rod-climbing (*Weissenberg effect*).
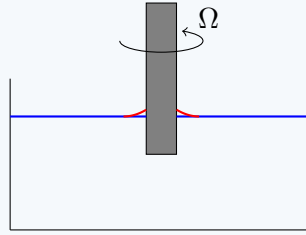
   Consider a vertical rod rotating at a constant rate Ω placed into a fluid. In a Newtonian fluid, viscous enough for Stokes equations to apply, there is no change in the position of the interface (blue).

   In a non-Newtonian fluid, the interface climbs up the rod (red). This is due to elastic tension: rotation creates circular streamlines. Tension along circles creates *hoop stress* which 'squeezes' the fluid and thus climbs up the rod.

2. Particle migration in a pipe flow.
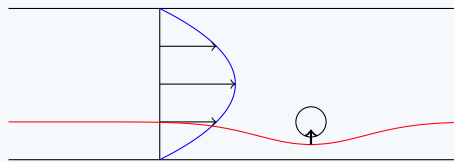
   In the case of Newtonian Stokes flow, the flow has no component perpendicular to the walls so a particle remains the same distance from the wall as it moves along the pipe. In the non-Newtonian case, hoop stress caused by curved streamlines lifts the particle away from the wall.

3.4 Extensional Viscosity

A shear flow is a *weak flow*: there is algebraic growth of distances between particles. In a *strong flow*, distances grow exponentially.

Consider a fluid with extension in the x and y directions and compression in the z direction: $\boldsymbol{u} + \dot{\varepsilon}\left(\frac{1}{2}x, \frac{1}{2}y, -z\right)$. We call $\dot{\varepsilon}$ the *extension rate*. Note that this flow is incompressible. The shear rate tensor is

$$\dot{\gamma} = \dot{\varepsilon}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}$$

In the Newtonian case, the shear rate tensor is

$$\sigma = \begin{pmatrix} -p_0 + \eta\dot{\varepsilon} & 0 & 0 \\ 0 & -p_0 + \eta\dot{\varepsilon} & 0 \\ 0 & 0 & -p_0 - 2\eta\dot{\varepsilon} \end{pmatrix}$$

Definition. The *extensional viscosity* is

$$\eta_{\text{ext}} = \frac{\sigma_{xx} - \sigma_{zz}}{\dot{\varepsilon}}$$

which has units of viscosity.

Thus for the Newtonian fluid, $\eta_{\text{ext}} = 3\eta$ which is constant.

Definition. The *Troutou ratio* is

$$\text{Tr} = \frac{\eta_{\text{ext}}}{\eta}$$

By the above calculations, Newtonian fluids have $\text{Tr} = 3$. Non-Newtonian fluids tend to have $\text{Tr} \gg 1$ in some range of shear rates. Thus complex fluids have a very different response to strong flows compared with weak flows.
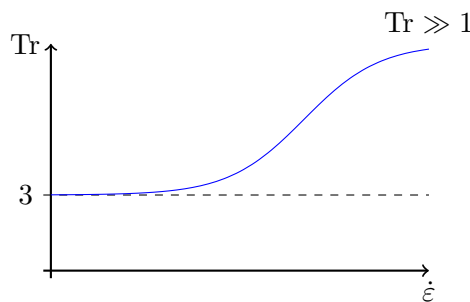
4 Generalised Newtonian Fluids

We will focus on steady flows and fluids which are *inelastic*. We will find how to incorporate shear-dependent viscosity into the constitutive relationship. One way is to generalise the Newtonian constitutive relationship to

$$\sigma = -p\mathbf{1} + \eta(\dot{\gamma})\dot{\gamma}$$

where $\eta(\dot{\gamma})$ is found empirically. These are called *generalised Newtonian fluids* (GNF).

We have a scalar function η of a tensor $\dot{\gamma}$, which is not in general coordinate invariant. Thus η must be a function of the *invariants* of $\dot{\gamma}$. A rank 2 tensor in 3 dimensions has 3 invariants. These are combinations of trace, determinants, and eigenvalues. We choose as the three invariants:

$$\text{tr}\,(\dot{\gamma})\,, \qquad \text{tr}\,(\dot{\gamma} \cdot \dot{\gamma})\,, \qquad \text{tr}\,(\dot{\gamma} \cdot \dot{\gamma} \cdot \dot{\gamma})$$

We always have $\text{tr}\,(\dot{\gamma}) = 0$ because the flow is incompressible:

$$\text{tr}\,(\dot{\gamma}) = \dot{\gamma}_{ii} = \frac{\partial u_i}{\partial x_i} + \frac{\partial u_i}{\partial x_i} = 2\nabla \cdot \boldsymbol{u} = 0$$

Consider a simple shear flow $\boldsymbol{u} = \dot{\gamma} y \hat{\boldsymbol{x}}$. Then

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

$$\dot{\gamma} \cdot \dot{\gamma} = \begin{pmatrix} \dot{\gamma}^2 & 0 & 0 \\ 0 & \dot{\gamma}^2 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$\dot{\gamma} \cdot \dot{\gamma} \cdot \dot{\gamma} = \begin{pmatrix} 0 & \dot{\gamma}^3 & 0 \\ \dot{\gamma}^3 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Thus $\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma} \cdot \dot{\gamma}) = 0$ and $\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma}) = 2\dot{\gamma}^2$. We assume all flows are approximately steady shear flow. Then $\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma})$ is the only non-zero invariant and $\eta(\dot{\gamma}) = \eta(\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma}))$.

> **Definition.** The *magnitude of shear rate* is
>
> $$\dot{\gamma} \equiv \left(\frac{\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma})}{2} \right)^{1/2} = \left(\frac{\dot{\gamma} : \dot{\gamma}}{2} \right)^{1/2}$$
>
> Note that $\dot{\gamma} \geq 0$ and for a simple shear flow the magnitude of shear rate $\dot{\gamma} = |\dot{\gamma}|$ which is the shear rate from steady shear flow. Thus the definitions coincide.

Note the second invariant $\text{tr}\,(\dot{\gamma} \cdot \dot{\gamma}) = \dot{\gamma} : \dot{\gamma} \geq 0$ and is zero only when there is no deformation (since $\dot{\gamma} : \dot{\gamma}$ is proportional to viscous dissipation), i.e. rigid body motion.

Recall for a simple shear flow, the shear stress is $\sigma = \eta(\dot{\gamma})\dot{\gamma}$, thus the above definitions agree with measurements of shear-dependent viscosity.

4.1 Power-law fluids

There are many choices for the function $\eta(\dot{\gamma})$ for a generalised Newtonian fluid. Here, we choose a *power-law*

$$\eta(\dot{\gamma}) \equiv \kappa \dot{\gamma}^{n-1}$$

where $\kappa > 0$ and $n \in \mathbb{Z}$ is the *power-index* of the fluid. Note $n = 1$ for a Newtonian fluid. For dimensional consistency, we require $[\kappa] = \text{Pa} \cdot \text{s}^n$.
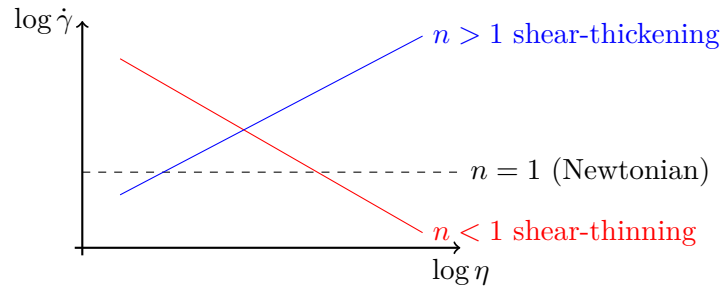
Note that in the limit $\dot{\gamma} \to 0$, we cannot define a zero-shear rate viscosity η_0 unless $n = 1$. Thus the model is problematic at small shear rates. The model is appropriate only for a finite range of shear rates.

Example. Newton's experiment with a power-law fluid. We assume there are no external pressures, and the flow is unidirectional: $\boldsymbol{u} = u(y)\hat{\boldsymbol{x}}$. We have

$$\dot{\gamma} = \begin{pmatrix} 0 & \frac{\partial u}{\partial y} & 0 \\ \frac{\partial u}{\partial y} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Then the magnitude of shear rate $\dot{\gamma} = \left| \frac{\partial u}{\partial y} \right|$. To find the flow, we use the Cauchy equation in 2D:

$$\frac{\partial p}{\partial y} = 0$$

$$\frac{\partial p}{\partial x} = \frac{\partial \sigma_{xy}}{\partial y}$$

since σ_{xy} is the only non-zero component of the shear stress. The first equation implies $p = p(x)$ only, which combined with the second implies $\sigma_{xy} = $ const.. Note we have not used the constitutive relationship yet: this is true for all GNFs. We have

$$\sigma_{xy} = \kappa \dot{\gamma}^{n-1} \dot{\gamma} = \kappa \dot{\gamma}^n = \kappa \left| \frac{\partial u}{\partial y} \right|^n = \text{const.}$$

Thus u_y is constant, i.e. u is linear in y, as with a Newtonian fluid.

If $\eta(\dot{\gamma})\dot{\gamma} = \sigma$ is a one-to-one function of $\dot{\gamma}$ then the result is the same: u varies linearly. A Couette flow $\boldsymbol{u} = Uy/h\hat{\boldsymbol{x}}$ is a *viscometric flow*: this flow is realised for all constitutive relationships provided σ is indeed a one-to-one function of $\dot{\gamma}$.

How do experimentally measure $\eta(\dot{\gamma})$? One method is a *shear flow rheometer*:

1. Impose $\dot{\gamma}$, measure σ (or vice versa)

2. Measure $\eta = \sigma/\dot{\gamma}$

3. Repeat varying $\dot{\gamma}$ or σ

4.1.1 Pipe flow of a power-law fluid

Consider axisymmetric pressure-driven flow in a pipe of a power-law GNF. If the fluid was Newtonian, we would get Poiseuille flow with a parabolic flow profile.
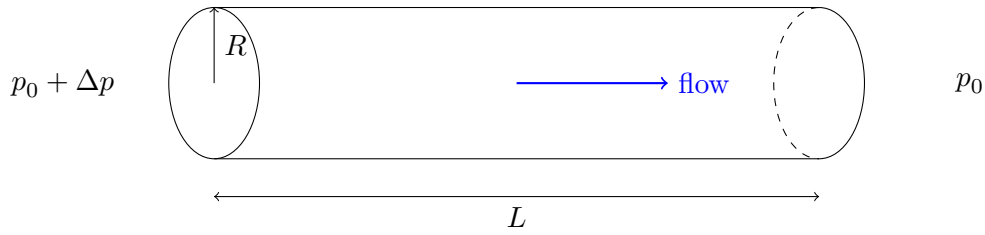
We will use cylindrical coordinates (r, θ, z) and assume the flow is unidirectional: $\boldsymbol{u} = u(r)\hat{\boldsymbol{z}}$. Then

$$\dot{\gamma} = \begin{pmatrix} 0 & 0 & \frac{\partial u}{\partial r} \\ 0 & 0 & 0 \\ \frac{\partial u}{\partial r} & 0 & 0 \end{pmatrix}$$

The magnitude of shear rate $\dot{\gamma} = \left| \frac{\partial u}{\partial r} \right|$. The Cauchy equations in cylindrical coordinates are

$$\frac{\partial p}{\partial r} = 0$$
$$\frac{\partial p}{\partial \theta} = 0$$
$$\frac{\partial p}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} (r \sigma_{rz})$$

since σ_{rz} is the only non-zero component of shear stress. The first two equations imply $p = p(z)$. Then $\frac{\partial p}{\partial z}$ is a function of z only, but the RHS is a function of r only. Thus each must be constant. Then

$$\frac{\partial p}{\partial z} = -\frac{\Delta p}{L} \implies \sigma_{rz} = -\frac{\Delta p}{2L} r + \frac{A}{r}$$

The A/r term is singular at $r = 0$ thus $A \equiv 0$. Note these results are true for all guids - we have not yet used the fact this is a power-law fluid.

Denote the *magnitude of wall shear stress* as σ_w. In this case, $\sigma_w = \frac{\Delta p}{2L} R$. To find the flow field, we need the constitutive relationship. For a GNF we have

$$\sigma_{rz} = \eta \left(\left| \frac{\partial u}{\partial r} \right| \right) \frac{\partial u}{\partial r} = -\frac{\Delta p}{2L} r$$

We expect u to be at maximum when $r = 0$, so expect $\frac{\partial u}{\partial r} < 0$. Thus $|u_r| = -u_r$. For a power-law fluid, $\sigma = \kappa \dot{\gamma}^n$, thus

$$\kappa \left| \frac{\partial u}{\partial r} \right|^n = \frac{\Delta p}{2L} r$$
$$\implies \left| \frac{\partial u}{\partial r} \right| = \left(\frac{\Delta p}{2L\kappa} \right)^{1/n} r^{1/n} = -\frac{\partial u}{\partial r}$$
$$\implies u(r) = C - \left(\frac{\Delta p}{2L\kappa} \right)^{1/n} \frac{n}{n+1} r^{\frac{n+1}{n}}$$

Enforcing no-slip boundary conditions on the pipe wall $u(R) = 0$ and re-writing in terms of the wall shear stress we have
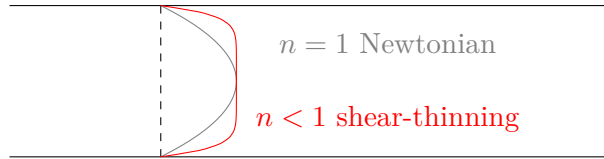
$$u(r) = \left(\frac{\sigma_w}{\kappa R} \right)^{1/n} \frac{n}{n+1} \left(R^{\frac{n+1}{n}} - r^{\frac{n+1}{n}} \right)$$

We can calculate the mean flow speed $\bar{U}$:

$$\bar{U} \equiv \frac{1}{\pi R^2} \iint u \, \mathrm{d}S = \frac{2}{R^2} \int_0^R r u(r) \, \mathrm{d}r = \left(\frac{\sigma_w}{\kappa}\right)^{1/n} \frac{nR}{3n+1}$$

Finally, we can rewrite the solution as flow relative to the mean flow speed:

$$\frac{u(r)}{\bar{U}} = \frac{3n+1}{n+1} \left[1 - \left(\frac{r}{R}\right)^{\frac{n+1}{n}}\right]$$

Physically, we have high shear near the pipe walls, so low viscosity in a shear-thinning complex fluid. The flow rate is

$$Q = \iint u \, \mathrm{d}S = \frac{\pi n}{3n+1} \left(\frac{\Delta p}{2L\kappa}\right)^{1/n} R^{3+\frac{1}{n}}$$

For a Newtonian fluid, $Q \sim \Delta p R^4$ whereas for a power-law fluid $Q \sim \Delta p^{1/n} R^{3+\frac{1}{n}}$. For a shear-thinning fluid with $n < 1$, we thus have a very strong dependence on Δp and R. In a device with Q fixed, $\Delta p^{1/n} R^{3+\frac{1}{n}} = \text{const.}$ so $\Delta p \sim R^{-(3n+1)}$. For $n < 1$, it is therefore easier to push fluid through a pipe than with a Newtonian fluid.

4.2 Other models & problems

4.2.1 Carreau-Yasuda model

The Carreau-Yasuda model can be written

$$\frac{\eta(\dot{\gamma}) - \eta_\infty}{\eta_0 - \eta_\infty} = [1 + (\lambda\dot{\gamma})^a]^{n-1} \, a$$

for $n \leq 1$. In this model, η transitions smoothly from η_0 to η_∞. In some finite range of shear rates, the model resembles a power-law fluid. In fact, a power-law fluid can be thought of as a 'subset' of Carreau-Yasuda. The constant parameter a is related to the curvature of the η vs. $\dot{\gamma}$ curve, whilst n is related to the steepness of the curve.
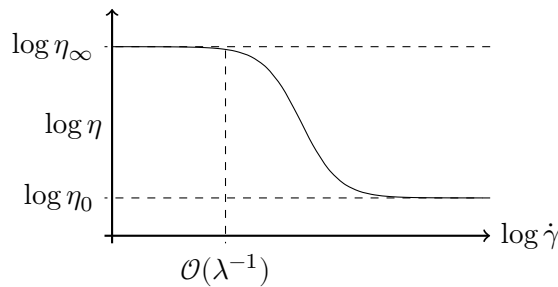
4.2.2 Powell-Eyring & Ellis models

The Powell-Eyring model is given by

$$\frac{\eta(\dot{\gamma}) - \eta_\infty}{\eta_0 - \eta_\infty} = \frac{\sinh^{-1}(\lambda\dot{\gamma})}{\lambda\dot{\gamma}}$$

Some models specify $\eta(\sigma)$ instead of $\eta(\dot{\gamma})$, for example the Ellis model.

$$\eta = \eta_0 \left[1 + \left| \frac{\sigma}{\sigma_0} \right|^{1-n} \right]^{-1}$$

There are several properties and issues associated with generalised Newtonian fluids.

1. Empirical: GNFs are based on experiments rather than derived.

2. Instantaneous: GNFs have no memory (recall the step shear experiment), so are not suitable for modelling unsteady flows. This also means GNFs are instantaneously reversible which is undesired.

3. No normal stress differences.

4. Under extension, the Troutou ratio is Tr = 3 for GNFs, i.e. there is no increase in extensional viscosity.

5. The behaviour of stress when $\dot{\gamma} \to 0$ is important. If η_0 can be defined, then $\sigma \to 0$. If not, such as with a power-law fluid, there is problematic behaviour near 0. The limit $\dot{\gamma} \to 0$ is often used to recover Newtonian behaviour.

4.3 Rheometry

Rheometry is the science and engineering of measuring material properties of fluids. For generalised Newtonian fluids, we need to measure $\eta(\dot{\gamma})$. The 'easy' method is to use a steady shear flow, which is simple in principle. However, we can only use a finite volume of fluid. A common solution is the *parallel plate rheometer*, also known as a parallel disc rheometer.

Consider two coaxial rigid circular discs of radius a held at a constant separation h. The bottom plate is held stationary whilst the upper plate rotates at a prescribed angular velocity Ω. The variable $\dot{\gamma}$ is controlled in this experiment.
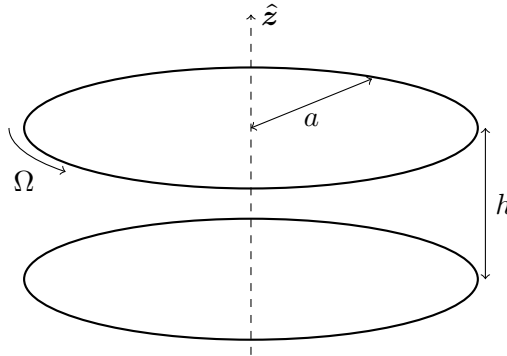
The input-output relationship can be inverted to infer $\eta(\dot\gamma)$. Use cylindrical coordinates (r, θ, z) and look for an axisymmetric flow field $\boldsymbol{u} = u(r, z)\widehat{\boldsymbol{\theta}}$. The shear rate tensor is

$$
\dot\gamma = \begin{pmatrix} 0 & r\frac{\partial}{\partial r}\left(\frac{u}{r}\right) & 0 \\ r\frac{\partial}{\partial r}\left(\frac{u}{r}\right) & 0 & \frac{\partial u}{\partial z} \\ 0 & \frac{\partial u}{\partial z} & 0 \end{pmatrix}
$$

Consider the steady Cauchy equation in cylindrical coordinates:

$$
\frac{\partial p}{\partial \theta} = 0 = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \tau_{r\theta}\right) + \frac{\partial}{\partial z}\left(\tau_{\theta z}\right)
$$

$$
\frac{\partial p}{\partial r} = 0
$$

$$
\frac{\partial p}{\partial z} = 0
$$

We wish to find a viscometric flow solution. In the Newtonian case, we have

$$
0 = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^3 \frac{\partial}{\partial r}\left(\frac{u}{r}\right)\right) + \frac{\partial^2 u}{\partial z^2}
$$

To find a solution we require both terms vanish. Thus $u = A(r)z + B(r)$. The no-slip boundary condition $u = 0$ at $z = 0$ and $u = \Omega r$ at $z = h$ gives

$$
u(r, z) = \Omega r \frac{z}{h}
$$

Given this flow, the shear rate tensor becomes

$$
\dot\gamma = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{r\Omega}{h} \\ 0 & \frac{r\Omega}{h} & 0 \end{pmatrix}
$$

Assume $\Omega > 0$, so the magnitude of the shear rate is $\dot\gamma = |r\Omega/h|$. This solution is also a solution for a GNF: we have $\tau_{r\theta} = 0$ and

$$
\tau_{\theta z} = \eta(\dot\gamma)\dot\gamma = \eta(\frac{r\Omega}{h})\frac{r\Omega}{h} = \tau_{\theta z}(r)
$$

The torque exerted to rotate the top plate is $\boldsymbol{T} = T\widehat{\boldsymbol{z}}$ where

$$
T = \int r\tau_{\theta z}\, \mathrm{d}S = 2\pi \int_0^a r^2 \eta(\dot\gamma)\dot\gamma\, \mathrm{d}r
$$

This relationship can be used to infer $\eta(\dot\gamma)$ by a change of variable. Let $\dot\gamma_a = a\Omega/h$ be the shear rate at the edge of the disc. Substitute $r = a\dot\gamma/\dot\gamma_a$:

$$
T = 2\pi \int_0^{\dot\gamma_a} \frac{a^2}{\dot\gamma_a^2}\dot\gamma^2 \eta(\dot\gamma)\dot\gamma \frac{a}{\dot\gamma_a}\, \mathrm{d}\dot\gamma = 2\pi \frac{a^3}{\dot\gamma_a^3} \int_0^{\dot\gamma_a} \eta(\dot\gamma)\dot\gamma^3\, \mathrm{d}\dot\gamma
$$

This is valid for all values of $\dot\gamma_a$. Rearranging and differentiating with respect to $\dot\gamma_a$ we have

$$
\eta(\dot\gamma_a) = \frac{1}{\dot\gamma_a^3}\frac{\mathrm{d}}{\mathrm{d}\dot\gamma^a}\left[\frac{T\dot\gamma_a^3}{2\pi a^3}\right]
$$

In an experiment, we control $\dot\gamma_a$, measure T, slightly change $\dot\gamma_a$, and repeat. Application of the above result then yields $\eta(\dot\gamma_a)$. Other geometries can also be used:

- Taylor-Couette.
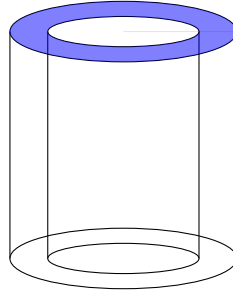
- Cone and plate. Useful since α small implies the shear rate $\dot{\gamma}$ is uniform.
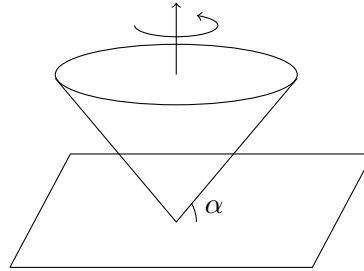
Lecture 5
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4.4 Variational approach

So far, we have found exact solutions only. For many GNF, no exact solutions are available so we must find approximate solutions. Here we consider a method relying on a form of energy minimisation. Conider a fluid volume V, bounded by a surface S, in the Stokes flow limit (no inertia). Assume $\boldsymbol{u} = \boldsymbol{u}_0$ is prescribed on S. The total rate of energy dissipation is

$$P = \int_V \frac{1}{2}\boldsymbol{\sigma} : \dot{\gamma}\,\mathrm{d}V = \int_V \frac{\eta}{2}\dot{\gamma} : \dot{\gamma}\,\mathrm{d}V \geq 0$$

Recall the minimum dissipation theorem: solutions to Stokes equations minimise P over all incompressible flows satisfying the same boundary conditions. The inequality $P_{\text{Stokes}} \leq P$ can be shown directly, or we can use the calculus of variations. Define a Lagrangian

$$\mathcal{L} \equiv P + \int_V \lambda \nabla \cdot \boldsymbol{u}\,\mathrm{d}V$$

Variations with respect to λ give $\nabla \cdot \boldsymbol{u} = 0$, and variations with respect to $\boldsymbol{u}$ give Stokes equations with pressure $p \propto \lambda$. To generalise this to GNF, consider the power density $f = \frac{1}{2}\eta\dot{\gamma}^2$ for a Newtonian fluid. Analogously, for a GNF we define

$$f = \int_0^{\dot{\gamma}} \eta(x)x\,\mathrm{d}x$$

and a new Lagrangian

$$\mathcal{L} \equiv \mathcal{L}\,[\boldsymbol{u}, \lambda] = \int_V f\,\mathrm{d}V + \int_V \lambda \nabla \cdot \boldsymbol{u}\,\mathrm{d}V$$

16

4.4.1 Calculus of variations

Consider a functional J of a vector field $\boldsymbol{y}(\boldsymbol{x})$ and its spatial derivatives $y_{m,n} \equiv \frac{\partial y_m}{\partial x_n}$;

$$J[\boldsymbol{y}] = \int_V F(x_i, y_j, y_{m,n}) \, \mathrm{d}V$$

where V is the total domain. The *first variation* of J is δJ where

$$J + \delta J = J[\boldsymbol{y} + \delta \boldsymbol{y}] = \int_V F(\boldsymbol{x}, \boldsymbol{y} + \delta \boldsymbol{y}) \, \mathrm{d}V$$

To determine an explicit expression for the first variation, first Taylor expand the integrand:

$$F(\boldsymbol{x}, \boldsymbol{y} + \delta \boldsymbol{y}) = F + \delta y_j \frac{\partial F}{\partial y_j} + \delta y_{m,n} \frac{\partial F}{\partial y_{m,n}} + \ldots$$

Using integration by parts and writing $\delta y_{m,n} = \frac{\partial}{\partial x_n} \delta y_m$ we have

$$\delta y_{m,n} \frac{\partial F}{\partial y_{m,n}} = \frac{\partial}{\partial x_n} \left(\delta y_m \frac{\partial F}{\partial y_{m,n}} \right) - \delta y_m \frac{\partial}{\partial x_n} \left(\frac{\partial F}{\partial y_{m,n}} \right)$$

The first term becomes a surface integral via Stokes' theorem, which may or may not vanish depending on boundary conditions. The first variation of F is thus

$$\delta F = \delta y_j \left[\frac{\partial F}{\partial y_j} - \frac{\partial}{\partial x_n} \left(\frac{\partial F}{\partial y_{j,n}} \right) \right] + \text{boundary terms}$$

Now requiring $\delta J = 0$ for all δy is equivalent to requiring $\delta F = 0$ for all δy, giving the *Euler-Lagrange equation*.

$$\frac{\partial F}{\partial y_j} - \frac{\partial}{\partial x_n} \left(\frac{\partial F}{\partial y_{j,n}} \right) = 0$$

4.4.2 Variational solution

Returning to our original set-up, denote $F_2 = \lambda \nabla \cdot \boldsymbol{u}$, $F_1 = \int_0^{\dot{\gamma}} \eta(x) x \, \mathrm{d}x$, so

$$\mathcal{L}[\boldsymbol{u}, \lambda] = \int_V F_1 + F_2 \, \mathrm{d}V$$

Note that we are using $\boldsymbol{y} \equiv \boldsymbol{u}$ here. The Euler-Lagrange equation for λ gives

$$\frac{\partial}{\partial \lambda} (F_1 + F_2) = 0 \implies \nabla \cdot \boldsymbol{u} = 0$$

For variations with respect to $\boldsymbol{u}$, we have boundary terms involving $\delta \boldsymbol{u}$. Since $\delta \boldsymbol{u} = \boldsymbol{0}$ on S, the boundary terms vanish in this case. There is no explicit dependence on $\boldsymbol{u}$ in F_1, F_2 so the Euler-Lagrange equation becomes

$$\frac{\partial}{\partial x_n} \left(\frac{\partial F}{\partial u_{j,n}} \right) = 0$$

where $F \equiv F_1 + F_2$. By chain rule,

$$\frac{\partial F_1}{\partial u_{j,n}} = \frac{\partial \dot{\gamma}}{\partial u_{j,n}} \frac{\partial F_1}{\partial \dot{\gamma}}$$

We have $\frac{\partial F_1}{\partial \dot{\gamma}} = \eta(\dot{\gamma})\dot{\gamma}$ and

$$
\begin{aligned}
\frac{\partial \dot{\gamma}}{\partial u_{j,n}} &= \frac{1}{2\dot{\gamma}} \frac{\partial}{\partial u_{j,n}} \left(\frac{1}{2} \dot{\gamma} : \dot{\gamma} \right) \\
&= \frac{1}{2\dot{\gamma}} \frac{\partial}{\partial u_{j,n}} \left(\frac{1}{2} \dot{\gamma}_{pq} \dot{\gamma}_{pq} \right) \\
&= \frac{1}{4\dot{\gamma}} \frac{\partial}{\partial u_{j,n}} \left(u_{p,q} u_{p,q} + 2 u_{p,q} u_{q,p} + u_{q,p} u_{q,p} \right) \\
&= \frac{1}{2\dot{\gamma}} \left(2 u_{j,n} + 2 u_{n,j} \right) \\
&= \frac{\dot{\gamma}_{jn}}{\dot{\gamma}}
\end{aligned}
$$

Now for $F_2 = \lambda \nabla \cdot \boldsymbol{u}$ we have

$$
\begin{aligned}
\frac{\partial}{\partial x_n} \left(\frac{\partial F_2}{\partial u_{j,n}} \right) &= \frac{\partial}{\partial x_n} \left(\lambda \frac{\partial u_{i,i}}{\partial u_{j,n}} \right) \\
&= \frac{\partial}{\partial x_n} \left(\lambda \delta_{ij} \delta_{in} \right) \\
&= \frac{\partial \lambda}{\partial x_j}
\end{aligned}
$$

Thus the full Euler-Lagrange equation for $\boldsymbol{u}$ is

$$
\frac{\partial}{\partial x_n} \left[\eta(\dot{\gamma}) \dot{\gamma}_{jn} \right] + \frac{\partial \lambda}{\partial x_j} = 0
$$

for $j = 1, 2, 3$. Given $\lambda = -p$, this becomes the Cauchy equation for a GNF.

$$
-\nabla p + \nabla \cdot \left[\eta(\dot{\gamma}) \dot{\gamma} \right] = \boldsymbol{0}
$$

Pressure is used as a Lagrange multiplier to enforce incompressibility. Note: if the boundary conditions are different, e.g. $\boldsymbol{\sigma} \cdot \boldsymbol{n}$ is prescribed, we must distinguish surfaces where $\boldsymbol{u}$ is described and surfaces where $\boldsymbol{\sigma} \cdot \boldsymbol{n}$ is prescribed, and add a surface integral to the Lagrangian (see Example Sheet 1).

To generate approximate solutions to the Cauchy GNF equations, we note that the closer the value of $\mathcal{L}$ to its minimum, the better the approximation. Thus

1. Use incompressible test functions $\boldsymbol{u}_\alpha$ where α is a parameter

2. Substitute into Lagrangian, minimise with respect to α

3. If the minimum of $\mathcal{L}$ is at $\alpha = \alpha^*$ then $\boldsymbol{u}_{\alpha^*}$ is an approximate solution

Integrals involved in $\mathcal{L}$ in general have to be evaluated numerically. Choice of test functions comes from physical intuition, computation, or experiment.

5 Yield-Stress Fluids

Some fluids only flow when subject to stress above some threshold. We call this threshold the *yield-stress*, denoted by σ_y. Fluids with such a property are called *yield-stress fluids* or *viscoplastic*

fluids. Examples are mayonnaise; jelly; peanut butter; mud; and hair gel. Graphically, σ_y is the non-zero $\dot{\gamma}$-intercept of $\sigma(\dot{\gamma})$. Beyond the yield-stress, the fluid may have Newtonian, shear-thinning, shear-thickening, or some other behaviour.

The simplest model of a yield-stress fluid is a *Bingham fluid* where the behaviour is Newtonian beyond the yield-stress. For $-\infty < \dot{\gamma} < \infty$ we have

$$\begin{cases} \dot{\gamma} = 0 & |\sigma| < \sigma_y \\ \sigma = \text{sgn}(\dot{\gamma})\sigma_y + \eta\dot{\gamma} & |\sigma| > \sigma_y \end{cases}$$

Alternatively, non-linear behaviour after yield is described by the *Herschel-Bulkley model* where the fluid has power-law behaviour after yield.

$$\begin{cases} \dot{\gamma} = 0 & |\sigma| < \sigma_y \\ \sigma = \text{sgn}(\dot{\gamma})\sigma_y + \kappa|\dot{\gamma}|^{n-1}\dot{\gamma} & |\sigma| > \sigma_y \end{cases}$$
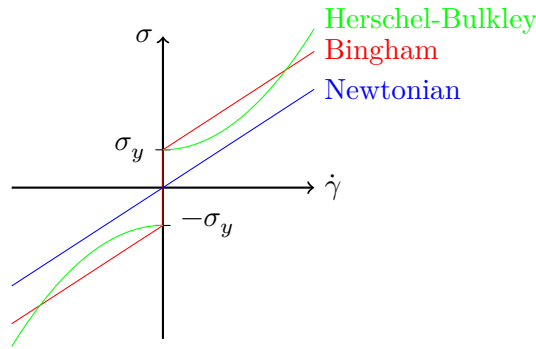
A typical flow problem of yield-stress fluids separates the domain into two regions.

1. Yielded domain: the fluid flows in regions where $|\sigma| > \sigma_y$

2. Unyielded domain: where $|\sigma| < \sigma_y$, there is no deformation i.e. rigid body motion only

The surface separating the yielded and unyielded regions is the *yield surface*. On this surface, $\sigma = \sigma_y$ by definition, which implicitly defines the surface. Physically, we expect high shear at boundaries, and thus expect the yield surface is close to these boundaries.

5.1 Pressure-driven flow of a Bingham fluid in a pipe

This problem illustrates a steady flow problem and the location of the yield surface. We will use cylindrical coordinates, and assume the flow is unidirectional with $\boldsymbol{u} = u(r)\hat{\boldsymbol{z}}$.
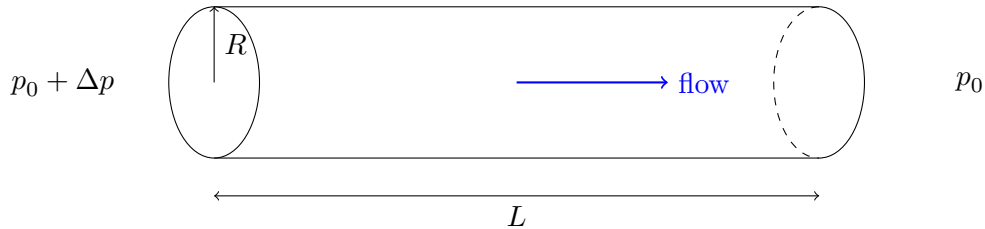
The Cauchy equation in cylindrical polars (as seen in section 4.1.1) gives

$$\frac{\partial p}{\partial z} = \frac{1}{r}\frac{\partial}{\partial r}(r\sigma_{rz}) = -\frac{\Delta p}{L}$$

thus as before we find

$$\sigma_{rz} = -\frac{\Delta p}{2L}r$$

We wish to find the yield surface. Clearly near $r = 0$ the stress is small so the fluid is unyielded. The yield surface is defined by $|\sigma| = \sigma_y$ thus we have

$$r = r_y = \frac{2L}{\Delta p}\sigma_y$$

Recall R is the radius of the pipe. Comparing R with r_y,

1. if $r_y \geq R$ the shear stress is below yield stress thoughout the pipe, thus there is no flow. Equivalently, this occurs for pressure differences $\Delta p \leq \frac{2L}{R}\sigma_y$.

2. if $r_y < R$ (equivalently $\Delta p > \frac{2L}{R}\sigma_y$) the fluid yields in $r_y \leq r \leq R$, and is unyielded in $0 \leq r \leq r_y$.

We solve case 2 exactly. Flow in the yielded region has $\dot{\gamma} = \frac{\partial u}{\partial r} < 0$. For a Bingham fluid $\sigma = \text{sgn}(\dot{\gamma})\sigma_y + \eta\dot{\gamma}$. Therefore we have

$$\sigma_{rz} = -\sigma_y + \eta\frac{\partial u}{\partial r}$$

Now $\sigma_{rz} = -\frac{\Delta p}{2L}r$ can be rewritten in terms of the yield surface as $\sigma_{rz} = -\sigma_y\frac{r}{r_y}$. Thus we must solve

$$\frac{\partial u}{\partial r} = \frac{\sigma_y}{\eta}\left(1 - \frac{r}{r_y}\right)$$

Note $r = r_y$ implies $\frac{\partial u}{\partial r} = 0$ which gives $|\sigma| = \sigma_y$ as expected. We have

$$u(r) = \frac{\sigma_y}{\eta}\left(r - \frac{r^2}{2r_y}\right) + c$$

The constant of integration is determined by the no-slip boundary condition on $r = R$, $u(R) = 0$. Finally we have for $r_y \leq r \leq R$

$$u(r) = \frac{\sigma_y}{\eta}\left(r - \frac{r^2}{2r_y} - R + \frac{R^2}{2r_y}\right)$$

In the unyielded region we have plug flow $u = $ constant. For $0 \leq r \leq r_y$, the rigid body velocity $u = U_y = u_{\text{yielded}}(r_y)$ by continuity.

$$\implies U_y = \frac{\sigma_y}{\eta}\left(\frac{r_y}{2} - R + \frac{R^2}{2r_y}\right) \geq 0$$

The flow profile in the pipe is a combination of parabolic and constant velocity.
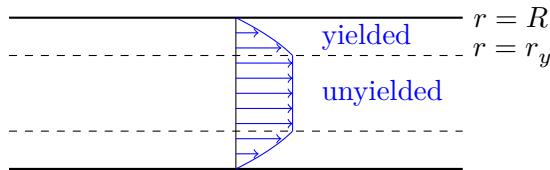
The flow rate Q vanishes if $r_y \geq R$. For $r_y < R$,

$$
\begin{aligned}
Q &= 2\pi \int_0^R r u(r) \, \mathrm{d}r \\
&= 2\pi \int_0^{r_y} r U_y \, \mathrm{d}r + 2\pi \int_{r_y}^R r u(r) \, \mathrm{d}r \\
&= \dots \\
&= \frac{\Delta p \pi R^4}{8 \mu L} \left[1 - \frac{4}{3} \frac{r_y}{R} + \frac{1}{3} \left(\frac{r_y}{R} \right)^3 \right]
\end{aligned}
$$

The constant factor outside the brackets is the *Hagen-Poiseuille flow rate* for a Newtonian fluid.

- If $r_y = 0$ the fluid is yielded everywhere and we have the Newtonian solution for Poiseuille flow in a pipe.

- If $r_y = R$ the fluid is unyielded everywhere and $Q = 0$.

- The term in brackets is always smaller than 1, thus $Q_{\text{yield-stress}} \leq Q_{\text{Newtonian}}$

This problem is doable because it is a steady problem. It is much more difficult in the unsteady case since the yield surface has unsteady motion, in which case inertia is needed.

5.2 Unsteady dynamics of yield surface

Here we include inertia to allow the yield surface to go from one steady state to another. Specifically, we consider a problem similar to Stokes' 1st problem, to illustrate the complexity of solving for the yield surface. In Stokes' 1st problem, a rigid plate at $y = 0$ impulsively starts to move at $t = 0$. The momentum 'diffuses' into the fluid as $y \sim \sqrt{\nu t}$.

Consider a Bingham fluid in $y > 0$ with yield stress σ_y, viscosity η, density ρ and a rigid plate at $y = 0$. For $t < 0$, we have a semi-infinite shear flow $\boldsymbol{u} = \dot{\gamma} y \hat{\boldsymbol{x}}$. Note: we require $\sigma = \eta \dot{\gamma} > \sigma_y$ to ensure there is flow everywhere. At $t = 0$, the plate starts to move with the flow such that the stress exerted on the plate suddenly changes to $\sigma_0 < \sigma_y$.
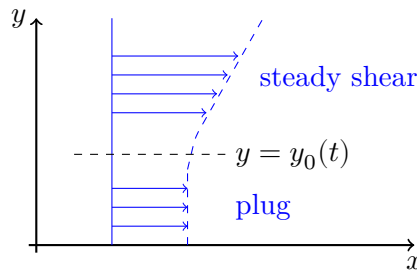
Physically, the unyielded domain appears near $y = 0$ and propagates into the yielded region. Let $y = y_0(t)$ be the yielded surface, so that $0 \leq y \leq y_0$ is the unyielded region region and $y > y_0$ is the unyielded region. On the yield surface, we require $|\sigma| = \sigma_y$ by definition, and also require the velocity to be continuous across the yield surface.

Assuming the flow is unidirectional with $\boldsymbol{u} = u(y, t) \hat{\boldsymbol{x}}$, the Cauchy equation with inertia is valid in both the yielded and unyielded domain:

$$
\rho \frac{\partial u}{\partial t} = \frac{\partial \sigma}{\partial y}
$$

Yielded region. We have $\sigma = \sigma_{xy} = \sigma_y + \eta \frac{\partial u}{\partial y}$, since $\dot{\gamma} = \frac{\partial u}{\partial y} \geq 0$. Thus the Cauchy equation becomes

$$\frac{\partial u}{\partial t} = \nu \frac{\partial^2 u}{\partial y^2}$$

where $\nu = \eta/\rho$ is the kinematic viscosity. This is a diffusion equation; as in Stokes' 1st problem, the yield surface has diffusive dynamics $y_0 \sim \sqrt{\nu t}$. Look for a similarity solution with dimensionless variable $s \equiv \frac{y}{\sqrt{\nu t}}$. Then

$$u(y,t) = \dot{\gamma} y f(s)$$
$$\frac{\partial u}{\partial t} = -\frac{1}{2} \frac{s}{t} \dot{\gamma} y \frac{\mathrm{d}f}{\mathrm{d}s}$$
$$\frac{\partial u}{\partial y} = \dot{\gamma} f + \dot{\gamma} s \frac{\mathrm{d}f}{\mathrm{d}s}$$
$$\frac{\partial^2 u}{\partial y^2} = 2 \frac{\dot{\gamma} s}{y} \frac{\mathrm{d}f}{\mathrm{d}s} + \dot{\gamma} \frac{s^2}{y} \frac{\mathrm{d}^2 f}{\mathrm{d}s^2}$$

The Cauchy equation is now

$$-\frac{1}{2} \frac{s}{t} \dot{\gamma} y \frac{\mathrm{d}f}{\mathrm{d}s} = \nu \left[2 \frac{\dot{\gamma} s}{y} \frac{\mathrm{d}f}{\mathrm{d}s} + \frac{\dot{\gamma} s^2}{y} \frac{\mathrm{d}^2 f}{\mathrm{d}s^2} \right]$$

Dividing by $\nu \dot{\gamma} s / y$ and rearranging we have the ODE

$$\left(2 + \frac{s^2}{2} \right) \frac{\mathrm{d}f}{\mathrm{d}s} + \frac{\mathrm{d}^2 f}{\mathrm{d}s^2} = 0$$

Since the ODE is second order, we require two boundary conditions:

1. Match steady shear as $y \to \infty$, i.e. $f(s) \to 1$.

2. At yield surface, $\sigma(y_0^+) = \sigma(y_0^-) = \sigma_y$. Equivalently, $u_y = 0$ at $y = y_0$. Thus we require

$$\dot{\gamma} f + \dot{\gamma} s \frac{\mathrm{d}f}{\mathrm{d}s} = 0$$

at $y = y_0$. Writing the location of the yield surface as $y_0 = \theta \sqrt{\nu t}$ with θ unknown we have

$$f(\theta) + \theta f'(\theta) = 0$$

Unyielded region. The same Cauchy equation holds, $\rho \partial_t u = \partial_y \sigma$. The flow in this region is a plug, so u does not depend on y. Thus $\partial_y \sigma$ is constant, i.e. σ is linear in y in this region. Now note at $y = y_0, \sigma = \sigma_y$ and at $y = 0, \sigma = \sigma_0$. So

$$\frac{\partial \sigma}{\partial y} = \frac{\sigma_y - \sigma_0}{y_0} = \frac{\sigma_y - \sigma_0}{\theta \sqrt{\nu t}}$$

Thus the Cauchy equation becomes

$$\rho \frac{\partial u}{\partial t} = \frac{\sigma_y - \sigma_0}{\theta \sqrt{\nu t}}$$

Integrating from $t' = 0$ to $t' = t$, we have

$$\rho u_{\text{unyielded}} = 2 \frac{\sigma_y - \sigma_0}{\theta \nu^{1/2}} t^{1/2}$$

Note $u_{\text{unyielded}} = u_{\text{yielded}}(y_0) = \dot{\gamma}y_0 f(\theta) = \dot{\gamma}\theta\sqrt{\nu t}f(\theta)$, so

$$\rho\dot{\gamma}\theta\sqrt{\nu t}f(\theta) = 2\frac{\sigma_y - \sigma_0}{\theta\nu^{1/2}}t^{1/2}$$
$$\implies \theta^2 f(\theta) = 2\xi$$

where $\xi = \frac{\sigma_y - \sigma_0}{\eta\dot{\gamma}}$ is a dimensionless control parameter, i.e. it is imposed on the system. This equation implicitly defines θ.

Yield surface.   Now the ODE for f gives

$$\frac{\mathrm{d}f}{\mathrm{d}s} = \frac{B}{s^2}e^{-\frac{s^2}{4}}$$

Integrating and using $f(s \to \infty) = 1$ we have

$$f(s) = 1 + \frac{C}{s}e^{-s^2/4} - \frac{\sqrt{\pi}}{2}C\mathrm{erfc}(s/2)$$

where $\mathrm{erfc}(x) \equiv \frac{2}{\sqrt{\pi}}\int_x^\infty e^{-u^2}\,\mathrm{d}u$ is the *complementary error function.* To determine C, we apply the BC $u_y = 0$ at the yield surface, i.e. $f(\theta) + \theta f'(\theta) = 0$

$$\implies C = \frac{2}{\sqrt{\pi}\mathrm{erfc}(\theta/2)}$$
$$\implies f(s) = 1 + \frac{\frac{2}{s}e^{-s^2/4} - \sqrt{\pi}\mathrm{erfc}(s/2)}{\sqrt{\pi}\mathrm{erfc}(\theta/2)}$$

The only remaining unknown is θ. Applying the condition $\theta^2 f(\theta) = 2\xi$, we have

$$\frac{\theta e^{-\theta^2/4}}{\sqrt{\pi}\mathrm{erfc}(\theta/2)} = \xi = \frac{\sigma_y - \sigma_0}{\eta\dot{\gamma}}$$

which is an implicit one-to-one equation for θ, so θ can be found numerically. Thus we have a full analytic solution up to an implicit equation for θ.

6 Linear Viscoelastic Fluids

Recall from phenomenology that some complex fluids have stresses which depend on the history of deformation, i.e. they have memory. Generalised Newtonian fluids and yield-stress fluids have no such memory. To model complex fluids with memory, we must incorporate *elasticity*.

6.1 Viscous vs. elastic

Consider fluids that have both a viscous *and* elastic response to deformation. Viscous response means that stress is proportional to rate of deformation, whilst an elastic response means stress is proportional to deformation. For example, polymer molecules are sheared and stretched by the flow, and provide an elastic response.

Recall Newton's experiment. Consider two points marked at the same x on the top and bottom plate at the same time, and the change in the mark as time passes. The change in position is d, the *shear displacement.*
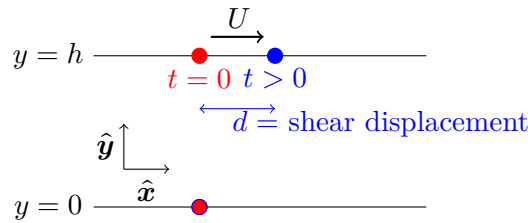
We then define *shear strain* $\gamma = \frac{d}{h}$, dimensionless, and as usual have shear rate $\dot{\gamma} = \frac{U}{h} = \frac{d\gamma}{dt}$. The viscous response (Newtonian) is $\sigma = \frac{F}{A} = \eta\dot{\gamma}$ where η is the viscosity. The *elastic* response, in analogy with a spring $F \propto kx$, we have $\sigma = G\gamma$ where G is the *shear modulus* and has units of stress.

Physically, a *dashpot* simulates viscous response, whilst a spring simulates elastic response. The simplest viscoelastic materials can be thought of as a linear combination of dashpots and springs.

6.2 Maxwell fluids

The most famous linear viscoelastic fluid (LVF) is a *Maxwell fluid*; modelled by a dashpot and a spring in *series*. We expect elastic behaviour on short timescales and viscous on long timescales. An example of a Maxwell fluid is a fluid with elastic inclusions such as droplets or vesicles.

1: spring, G 2: dashpot, η
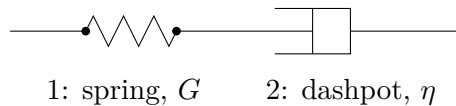
We can derive the constitutive relationship from this model. We have

$$\sigma_1 = G\gamma_1 \qquad \text{spring}$$
$$\sigma_2 = \eta\dot{\gamma}_2 \qquad \text{dashpot}$$

The total stress and strain follow by analogy with circuits (cf. Kirkhoff's laws). Strain is analogous to potential difference and stress is analogous to current. Therefore

$$\text{total strain} \quad \gamma = \gamma_1 + \gamma_2$$
$$\text{total stress} \quad \sigma = \sigma_1 = \sigma_2$$

The total rate of strain is

$$\dot{\gamma} = \dot{\gamma}_1 + \dot{\gamma}_2 = \frac{\dot{\sigma}_1}{G} + \frac{\sigma_2}{\eta} = \frac{\dot{\sigma}}{G} + \frac{\sigma}{\eta}$$
$$\implies \sigma + \frac{\eta}{G}\dot{\sigma} = \eta\dot{\gamma}$$

This is the constitutive relationship for a Maxwell fluid. $\lambda \equiv \frac{\eta}{G}$ is the *relaxation timescale* for stress.

Intepretation of λ**.** Consider a sudden cessation of deformation, that is

$$\begin{cases} \dot{\gamma} = 0 & t \geq 0 \\ \sigma = \sigma_0 & t < 0 \end{cases}$$

Thus for $t > 0$ we have $\sigma + \lambda\dot{\sigma} = 0$ with boundary conditions $\sigma(t = 0) = \sigma_0$. Thus

$$\sigma(t) = \sigma_0 e^{-\frac{t}{\lambda}}$$

Hence we have exponential relaxation of stress on timescale λ. Note λ is the timescale for adjustments in stress due to changes in deformation, whilst deformation adjusts instantaneously to changes in stress.

Behaviour at short vs. long timescales. Consider a Maxwell fluid for $t \ll \lambda$ and $t \gg \lambda$.

- On short timescales $t \ll \lambda$, we have $\lambda\dot{\sigma} \gg \sigma$ hence the Maxwell fluid has $\lambda\dot{\sigma} \sim \eta\dot{\gamma}$. This is an elastic response.

- On long timescales $t \gg \lambda$, we have $\lambda\dot{\sigma} \ll \sigma$ so the Maxwell fluid has $\sigma \sim \eta\dot{\gamma}$ which is a viscous response.

6.3 Kelvin-Voigt fluid

Here we consider a similar idea to introduce a fluid with the opposite behaviour to a Maxwell fluid. Consider a spring and dashpot in *parallel*.

1: spring, G
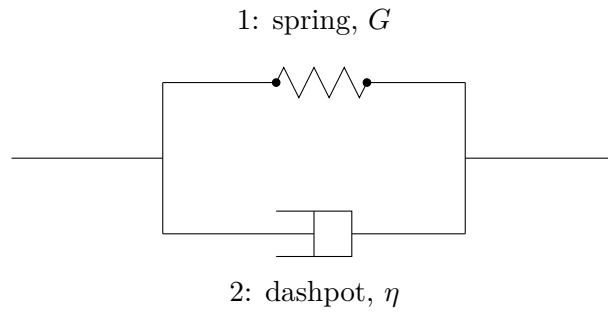
2: dashpot, η

We have individual stresses

$$\sigma_1 = G\gamma_1 \qquad \text{spring}$$
$$\sigma_2 = \eta\dot{\gamma}_2 \qquad \text{dashpot}$$

as before. In parallel, the total strain is $\gamma = \gamma_1 + \gamma_2$ and the total stress is

$$\sigma = \sigma_1 + \sigma_2 = G\gamma_2 + \eta\dot{\gamma}_2 = G\left[\gamma + \frac{\eta}{G}\dot{\gamma}\right]$$

Hence the constitutive relationship for a Kelvin-Voigt fluid is

$$\sigma = G\left[\gamma + \hat{\lambda}\dot{\gamma}\right]$$

where $\hat{\lambda}$ is the *retardation timescale*, the timescale for deformation to adjust to changes in stress. Note: stress adjusts instantaneously to changes in deformation in this case.

- On short timescales, we have $\hat{\lambda}\dot{\gamma} \gg \gamma$ so $\sigma \sim G\hat{\lambda}\dot{\gamma}$ is a viscous response.

- On long timescales, we have $\hat{\lambda}\dot{\gamma} \ll \gamma$ so $\sigma \sim G\gamma$ which is an elastic response.

The properties of Kelvin-Voigt fluids are opposite to those of a Maxwell fluid. Since a Kelvin-Voigt fluid behaves as an elastic material at long times, it is also referred to as a viscoelastic *material* or *solid*.

6.4 Jeffreys fluid

We wish to construct a LVF that has a viscous response at both *short* and *long* timescales. Thus we require two distinct timescales. A Jeffreys fluid has a dashpot and Kelvin-Voigt fluid in series.
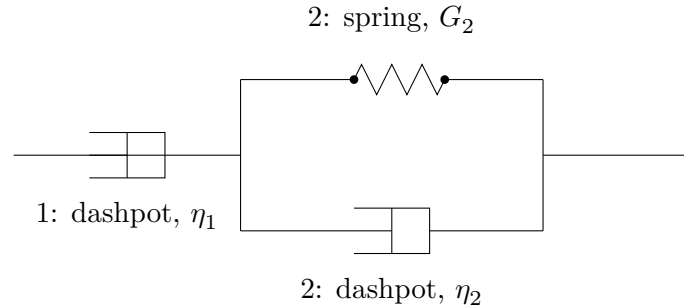
The total strain is $\gamma = \gamma_1 + \gamma_2$ where γ_1 is the strain of the lone dashpot and γ_2 is the total strain of the Kelvin-Voigt fluid. The total stress is $\sigma = \sigma_1 = \sigma_2$. We have

$$\sigma = \sigma_1 = \eta_1 \dot{\gamma}_1$$

$$\sigma = \sigma_2 = G_2 \gamma_2 + \eta_2 \dot{\gamma}_2 = \left[G_2 + \eta_2 \frac{\partial}{\partial t} \right] \gamma_2$$

To find the constitutive relationship, first differentiate the total strain:

$$\dot{\gamma} = \dot{\gamma}_1 + \dot{\gamma}_2 = \frac{\sigma}{\eta_1} + \dot{\gamma}_2$$

Now apply the operator $\left[G_2 + \eta_2 \frac{\partial}{\partial t} \right]$.

$$[G_2 + \eta_2 \partial_t] \dot{\gamma} = \frac{1}{\eta_1} [G_2 + \eta_2 \partial_t] \sigma + \partial_t [(G_2 + \eta_2 \partial_t) \gamma_2]$$

$$= \frac{G_2}{\eta_1} \sigma + \left(\frac{\eta_2}{\eta_1} + 1 \right) \dot{\sigma}$$

Thus the constitutive relationship for a Jeffreys fluid is

$$\left(1 + \lambda_1 \frac{\partial}{\partial t} \right) \sigma = \eta_1 \left(1 + \lambda_2 \frac{\partial}{\partial t} \right) \dot{\gamma}$$

where the timescales are

$$\text{relaxation timescale} \quad \lambda_1 = \frac{\eta_1 + \eta_2}{G_2}$$

$$\text{retardation timescale} \quad \lambda_2 = \frac{\eta_2}{G_2}$$

The fluid has both relaxation and retardation, with $\lambda_1 \geq \lambda_2$. Typically, λ_1 / λ_2 could be large. If $\lambda_2 = 0$ we have a Maxwell fluid.

- On short timescales $t \ll \lambda_1, \lambda_2$ we have $\lambda_1 \dot{\sigma} \sim \eta_1 \lambda_2 \partial_t \dot{\gamma} \implies \sigma \sim \eta_1 \frac{\lambda_2}{\lambda_1} \dot{\gamma}$ which is a viscous response.

- On long timescales $t \gg \lambda_1, \lambda_2$ we have $\sigma \sim \eta_1 \dot{\gamma}$ which is a viscous response.

- On intermediate timescales $\lambda_2 \ll t \ll \lambda_1$ we have $\lambda_1 \dot{\sigma} \sim \eta_1 \dot{\gamma} \implies \sigma \sim \frac{\eta_1}{\lambda_1} \dot{\gamma}$ which is an elastic response.

Lecture 9
5/11/20

6.5 General framework

What is the most general constitutive relationship for a linear viscoelastic fluid? The constitutive relationship for a *general linear viscoelastic fluid* (GLVF) can be written as

$$\sigma(t) = \int_{-\infty}^{t} G(t-t')\dot{\gamma}(t')\,\mathrm{d}t'$$

where $G(s)$ is the *relaxation modulus* of the fluid. G is a positive function which is decreasing in s, reflecting the fact memory fades over time. G has dimensions of stress. By causality, $G(s) = 0$ for $s < 0$ so that future changes in shear rate do not influence the present stress.

- A Newtonian fluid has $G_{\mathrm{N}}(s) = 2\eta\delta(s)$, using the convention that

$$\int_{\infty}^{t} \delta(t-t')\,\mathrm{d}t' = \frac{1}{2}$$

- A Maxwell fluid's relaxation modulus can be calculated from its constitutive relationship.

$$\sigma + \lambda\dot{\sigma} = \eta\dot{\gamma}$$

$$\implies \lambda\frac{\mathrm{d}}{\mathrm{d}t}\left[\sigma e^{t/\lambda}\right] = \eta\dot{\gamma}e^{t/\lambda}$$

$$\sigma(t) = \int_{\infty}^{t} \frac{\eta}{\lambda}\dot{\gamma}(t')e^{\frac{t'-t}{\lambda}}\,\mathrm{d}t'$$

  This is the Maxwell fluid constitutive relationship in integral form. Hence

$$G_{\mathrm{M}}(s) = \frac{\eta}{\lambda}e^{-s/\lambda}$$

  which shows a Maxwell fluid has an exponential loss of memory. Note that η has units of stress $\times$ time, and λ has units of time. Thus G has units of stress as expected.

- A *generalised* Maxwell fluid has relaxation modulus

$$G_{\mathrm{GMF}}(s) = \sum_{i=1}^{N} \frac{\eta_i}{\lambda_i}e^{-\frac{s}{\lambda_i}}$$

Generalising the scalar relationship, we get a tensorial relationship for deviatoric stress

$$\tau(t) = \int_{-\infty}^{t} G(t-t')\dot{\gamma}(t')\,\mathrm{d}t'$$

Note that G is still a scalar.

Steady flow. Consider a steady flow with $\dot{\gamma}(t') = \dot{\gamma}$ a constant tensor. Then

$$\tau(t) = \dot{\gamma}\int_{\infty}^{t} G(t-t')\,\mathrm{d}t' = \left[\int_{0}^{\infty} G(s)\,\mathrm{d}s\right]\dot{\gamma}$$

which is the Newtonian constitutive relationship. Thus if the flow is steady, stress is Newtonian with $\tau = \eta\dot{\gamma}$ and $\eta = \int_{0}^{\infty} G(s)\,\mathrm{d}s$. Hence Newtonian solutions to previous problems are the same for a GLVF if the flow is steady. To see the consequence of viscoelastic behaviour, we require unsteady motion where unsteady boundary conditions and inertia are involved.
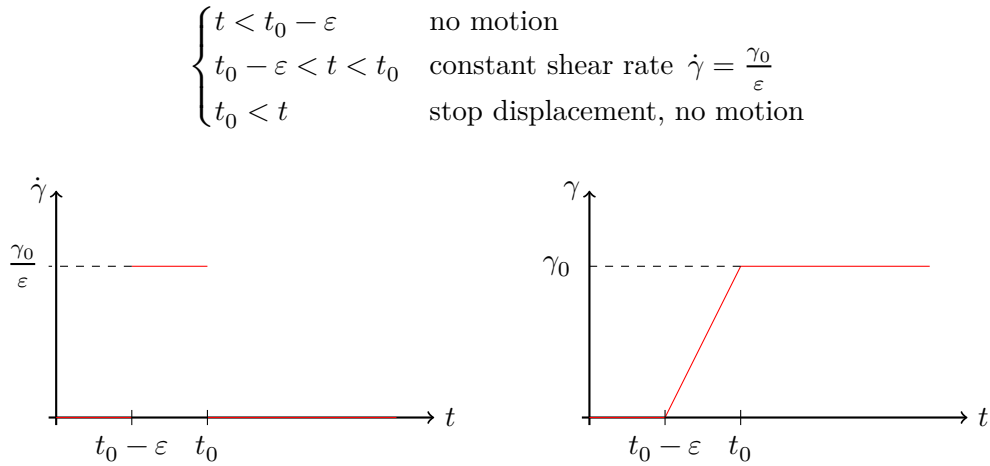
6.6 Interpretation of relaxation modulus

All experiments (shear flows and others) with a sudden start/stop require consideration of inertia. We wish to find a limit in which we can neglect inertia. Consider a simple shear flow setup, with sudden motion of one of the plates. The steady state is reached on a timescale $\tau \sim \dfrac{h^2}{\nu}$ where h is the gap between the plates and $\nu = \dfrac{\eta}{\rho}$ is the kinematic viscosity. Typical values of ν are $\nu \sim 10^{-6} m^2 s^{-1}$ for water and $\nu \sim 10^{-3} m^2 s^{-1}$ for glycerol. If τ is much smaller than any of the elastic timescales (e.g. relaxation or retardation times) then we can neglect inertia.

Example.

- Consider glycerol in a setup with $h \sim 100 \mu m, \tau \sim 10^{-5} s$. Then we can neglect inertia if $10^{-5} s \ll \lambda$.

- Consider a cone and plate geometry with small angle β. Typically $R \sim 1cm, h \sim 100 \mu m$. Water has $\nu \sim 10^{-6} m^2 s^{-1}$ so $\tau \sim 10^{-2} s$ and we can neglect inertia if $10^{-2} s \ll \lambda$.

Within this assumption, we can interpret $G(s)$. Consider stress relaxation after a sudden (short) shearing displacement.

$$\begin{cases} t < t_0 - \varepsilon & \text{no motion} \\ t_0 - \varepsilon < t < t_0 & \text{constant shear rate } \dot{\gamma} = \frac{\gamma_0}{\varepsilon} \\ t_0 < t & \text{stop displacement, no motion} \end{cases}$$

The shear stress is

$$\sigma = \sigma(t) = \int_{-\infty}^{t} G(t - t')\dot{\gamma}(t')\,\mathrm{d}t'$$

$$= \int_{t_0 - \varepsilon}^{t_0} G(t - t')\dot{\gamma}(t')\,\mathrm{d}t'$$

$$= \frac{\gamma_0}{\varepsilon} \int_{t_0 - \varepsilon}^{t_0} G(t - t')\,\mathrm{d}t'$$

In the limit $\varepsilon \to 0$ we get

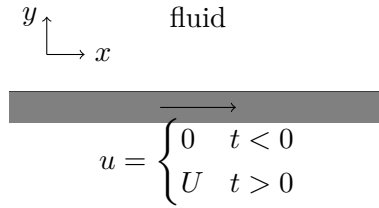$$\sigma(t) \to \gamma_0 G(t - t_0) \text{ as } \varepsilon \to 0$$

Thus the interpretation is that G is the (scaled) stress response to instantaneous shear displacement.

6.7 Stokes' 1st problem

Consider an impulsively started plate in a semi-infinite fluid. In the Newtonian case, this problem is classically used to gain intutition about diffusion of vorticity and momentum. Suppose the flow is unidirectional with $\boldsymbol{u} = u(y)\hat{\boldsymbol{x}}$. Then from the Cauchy equation we have

$$\rho\frac{\partial u}{\partial t} = \eta\frac{\partial^2 u}{\partial y^2}$$

The solution is a *similarity solution* with $u = Uf(y, \nu, t) = Uf(\frac{y}{\sqrt{\nu t}})$. Thus the diffusion dynamics reach $y \sim \sqrt{\nu t}$ at time t.

A Maxwell fluid with relaxation timescale λ instead has similarity solution

$$u = Uf(y, \nu, t, \lambda) = Uf(\frac{y}{\sqrt{\nu t}}, \frac{t}{\lambda})$$

This is in fact significantly more complicated. Equations satisfied by $u(y, t)$ are now

$$\sigma + \lambda\frac{\partial\sigma}{\partial t} = \eta\dot{\gamma} = \eta\frac{\partial u}{\partial y} \qquad\qquad \text{constitutive relationship}$$

$$\rho\frac{\partial u}{\partial t} = \frac{\partial\sigma}{\partial y} \qquad\qquad \text{Cauchy equation}$$

Applying the operator $[1 + \lambda\partial_t]$ to the Cauchy equation gives

$$\frac{\partial u}{\partial t} + \lambda\frac{\partial^2 u}{\partial t^2} = \nu\frac{\partial^2 u}{\partial y^2}$$

This can be solved explicitly via Fourier transforms. Instead, we will consider short and long timescales.

- Short times $t \ll \lambda$ give $\lambda\ddot{u} \gg \dot{u}$, hence

$$\lambda\frac{\partial^2 u}{\partial t^2} \approx \nu\frac{\partial^2 u}{\partial y^2}$$

   This is a wave equation for u with wavespeed $c = \sqrt{\nu/\lambda}$. For glycerol, $\nu \sim 10^{-3}m^2s^{-1}$ and $\lambda \sim 1s$ so $c \sim 1cms^{-1}$.

- Long times $t \gg \lambda$ give $\lambda\ddot{u} \ll \dot{u}$ hence

$$\frac{\partial u}{\partial t} = \nu\frac{\partial^2 u}{\partial y^2}$$

   This is a diffusion equation for u as in the Newtonian case.

- Transition times $t \sim \lambda$: there is a finite extent Δ of fluid subject to wave dynamics, with

$$\Delta \sim ct \sim c\lambda = \sqrt{\nu\lambda}$$

For glycerol, $\Delta \sim 1cm$. Δ increases with λ and η, and is independent of U as expected by linearity.

Suppose instead we have a GLVF. In this case, u satisfies

$$\sigma = \sigma(y,t) = \int_{-\infty}^{t} G(t-t')\dot{\gamma}(y,t')\,\mathrm{d}t' = \int_{-\infty}^{t} G(t-t')\frac{\partial u}{\partial y}(y,t')\,\mathrm{d}t'$$

Then the Cauchy equation $\rho u_t = \sigma_y$ gives

$$\rho\frac{\partial u}{\partial t} = \int_{-\infty}^{t} G(t-t')\frac{\partial^2 u}{\partial y^2}(y,t')\,\mathrm{d}t'$$

Lecture 10
10/11/20

6.8 Oscillatory Rheology

Recall *rheology* is the science of measuring the mechanical properties of complex fluids. The 'devices' used are *rheometers*. For example, a Couette device is used for simple shear flow. This devices requires large strains which is a common problem for rheometers. We can avoid this constraint by considering oscillatory rheometers, which only require small strains. There are four classical ways to implement such a device.

1. Couette device – uniform shear
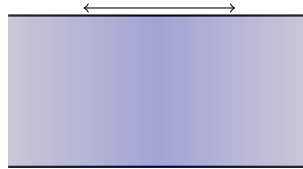
2. Parallel discs – non-uniform shear
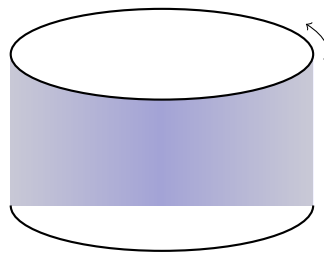
3. Taylor-Couette device – non-uniform shear

4. Cone-and-plate – approximately uniform shear

Consider oscillation with shear rate $\dot\gamma = \dot\gamma_0 \cos\omega t$ where γ_0 is the amplitude of shear strain and $\dot\gamma_0 = \gamma_0\omega$. The shear stress for a GLVF is then

$$\sigma = \sigma(t) = \int_{-\infty}^{t} G(t-t')\dot\gamma_0 \cos(\omega t')\,\mathrm{d}t'$$

$$\stackrel{s=t-t'}{=} \dot\gamma_0 \int_0^\infty G(s)\cos(\omega(t-s))\,\mathrm{d}s$$
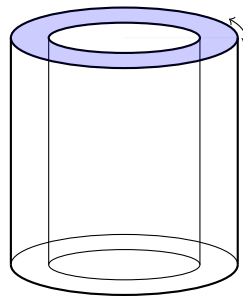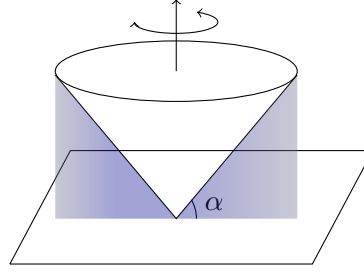
$$= \dot\gamma_0 \cos\omega t \left[\int_0^\infty G(s)\cos\omega s\,\mathrm{d}s\right]$$

$$+ \dot\gamma_0 \sin\omega t \left[\int_0^\infty G(s)\sin\omega s\,\mathrm{d}s\right]$$

where the first term is in phase with the shear rate, and the second term is in phase with the deformation. We can rewrite the shear stress as

$$\sigma = \gamma_0 \left[G'(\omega)\sin\omega t + G''(\omega)\cos\omega t\right]$$

where

$$G'(\omega) = \omega \int_0^\infty G(s)\sin\omega s\,\mathrm{d}s \qquad\qquad \text{storage modulus}$$

$$G''(\omega) = \omega \int_0^\infty G(s)\cos\omega s\,\mathrm{d}s \qquad\qquad \text{loss modulus}$$

The *storage modulus* is analogous to the storage of elastic potential energy in a spring, i.e. this component of the stress is a result of elastic behaviour. Similarly, the loss modulus is analogous to the dissipation of energy due to friction and is a result of viscous behaviour.

- The term in G' has $\sigma \propto \gamma = \gamma_0 \sin\omega t$ and hence G' is interpreted as an elastic constant, analogous to k for a spring.

- The term in G'' has $\sigma \propto \dot\gamma = \gamma_0\omega \cos\omega t$. Note that

$$\gamma_0 G''(\omega)\cos\omega t = \dot\gamma_0 \frac{G''(\omega)}{\omega}\cos\omega t$$

so we interpret G''/ω as a viscosity and define

$$\eta'(\omega) = \frac{G''(\omega)}{\omega} = \int_0^\infty G(s)\cos\omega s\,\mathrm{d}s$$

Finally, with these definitions, we have

$$\sigma = \gamma_0 G'(\omega)\sin\omega t + \dot{\gamma}_0 \eta'(\omega)\cos\omega t$$

Note if the fluid is Newtonian, $G' = 0$ and $\eta' = \eta$. We can write G' and G'' in a complex formulation:

$$G^*(\omega) = G'' + iG' = \omega \int_0^\infty G(s)e^{i\omega s}\,\mathrm{d}s$$
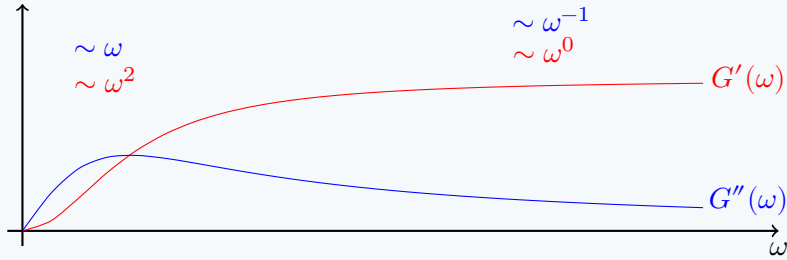
hence $G^*(\omega)$ is proportional to the Fourier Transform of $G(s)$.

Example. Consider a Maxwell fluid with $G(s) = \frac{\eta}{\lambda}e^{-s/\lambda}$. Then

$$\begin{aligned}
G^*(\omega) &= \omega \int_0^\infty \frac{\eta}{\lambda}e^{-s/\lambda}e^{i\omega s}\,\mathrm{d}s \\
&= \frac{\eta\omega}{\lambda}\int_0^\infty e^{(i\omega - \frac{1}{\lambda})s}\,\mathrm{d}s \\
&= -\frac{\eta\omega}{\lambda}\frac{1}{i\omega - \frac{1}{\lambda}} \\
&= \frac{\eta\omega}{1 - i\lambda\omega}
\end{aligned}$$

Taking real and imaginary parts we have

$$G'(\omega) = \Im[G^*] = \frac{\eta\lambda\omega^2}{1 + (\lambda\omega)^2}$$

$$G''(\omega) = \Re[G^*] = \frac{\eta\omega}{1 + (\lambda\omega)^2}$$

The maximum of $G''(x) = \frac{1}{1+x^2}$ occurs at $x = 1$ hence the maximum of $G''(\omega)$ is at $\lambda\omega = 1$. Thus at maximum, $\omega = \frac{1}{\lambda}$ which can be used to estimate λ experimentally. We can define a new dimensionless number, the *Deborah number* $\lambda\omega = \mathrm{De} = \frac{\lambda}{\omega^{-1}}$ which is a ratio of two timescales. If $\mathrm{De} \ll 1$, then $\lambda \ll \omega^{-1}$ and the Maxwell fluid has short memory, so an approximately Newtonian response. If $\mathrm{De} \gg 1$, then $\lambda \gg \omega^{-1}$ and there are strong elastic effects.

6.9 Further properties

The generalised linear visco-elastic fluid model allows modelling of fluids with memory and history-dependent shear rate / stress relationships. However, there are four main issues with LVFs:

1. *Constant viscosity in steady flow*, so the viscosity is independent of shear rate and we cannot model shear-thinning or thickening behaviour. Recall we found

$$\tau = \dot{\gamma}\eta$$

where $\eta = \int_0^\infty G(s)\,\mathrm{d}s$.

2. *No normal stress differences.* Recall steady shear flow has

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

$$\sigma = \begin{pmatrix} -p_0 & \eta\dot{\gamma} & 0 \\ \eta\dot{\gamma} & -p_0 & 0 \\ 0 & 0 & -p_0 \end{pmatrix}$$

Thus $N_1 = N_2 = 0$.

3. *No change in extensional viscosity.* Recall a steady extensional flow has

$$\dot{\gamma} = \dot{\varepsilon}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}$$

$$\sigma = \begin{pmatrix} -p_0 + \eta\dot{\varepsilon} & 0 & 0 \\ 0 & -p_0 + \eta\dot{\varepsilon} & 0 \\ 0 & 0 & -p_0 - 2\eta\dot{\varepsilon} \end{pmatrix}$$

Hence $\eta_{\text{ext}} = 3\eta$ and $\mathrm{Tr} = 3$ as for a Newtonian fluid.

4. The GLVF constitutive relationship is *not objective*: it depends on the frame in which it is evaluated.

6.10 Frame dependence

We have two issues to solve: translation of frame, and rotation of frame. To solve the issue of frame translation independence, we use familiar tools from continuum mechanics. To enforce Galilean invariance, we will use *material derivatives* instead of partial time derivatives, i.e. replace ∂_t with $\frac{\mathrm{D}}{\mathrm{D}t} = \partial_t + \boldsymbol{u} \cdot \nabla$.

6.10.1 Translation

Consider two frames of reference. Frame 1 is stationary, whilst frame 2 moves relative to frame 1 with constant velocity $\boldsymbol{U}$. Then

$$\boldsymbol{x}^{(1)} = \boldsymbol{x}^{(2)} + \boldsymbol{U}t$$
$$\boldsymbol{u}^{(1)} = \boldsymbol{u}^{(2)} + \boldsymbol{U}$$

The stress in frame 1 and 2 may be written

$$\sigma^{(1)} = \sigma^{(1)}(\boldsymbol{x}^{(1)}, t)$$
$$\sigma^{(2)} = \sigma^{(2)}(\boldsymbol{x}^{(2)}, t)$$

Now from Galilean invariance we have

$$\sigma(\boldsymbol{x}^{(2)}, t) = \sigma^{(1)}(\boldsymbol{x}^{(1)}, t) = \sigma^{(1)}(\boldsymbol{x}^{(2)} + \boldsymbol{U}t, t)$$

Therefore the material derivative of $\sigma^{(2)}$ is

$$
\begin{aligned}
\frac{\mathrm{D}\sigma^{(2)}}{\mathrm{D}t} &= \frac{\partial \sigma^{(2)}}{\partial t} + \boldsymbol{u}^{(2)} \cdot \nabla \sigma^{(2)} \\
&= \frac{\partial}{\partial t}\left[\sigma^{(1)}(\boldsymbol{x}^{(2)} + \boldsymbol{U}t, t)\right] + \boldsymbol{u}^{(2)} \cdot \nabla \sigma^{(2)} \\
&= \frac{\partial \sigma^{(1)}}{\partial t} + \boldsymbol{U} \cdot \nabla \sigma^{(1)} + \left(\boldsymbol{u}^{(1)} - \boldsymbol{U}\right) \cdot \nabla \sigma^{(1)} \\
&= \frac{\mathrm{D}\sigma^{(1)}}{\mathrm{D}t}
\end{aligned}
$$

Hence the material derivative is independent of the frame translation velocity $\boldsymbol{U}$. A Maxwell fluid previously had constitutive relationship $\sigma + \lambda \frac{\partial \sigma}{\partial t} = \eta \dot{\gamma}$. Now, we have
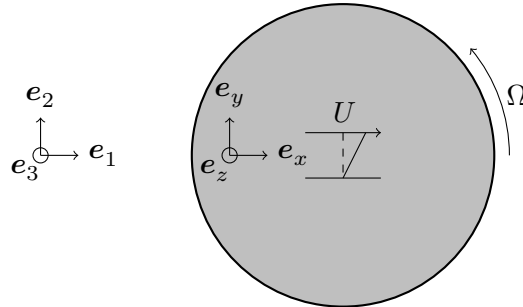
$$\sigma + \lambda \frac{\mathrm{D}\sigma}{\mathrm{D}t} = \eta \dot{\gamma}$$

Lecture 11
12/11/20

However, this is still dependent on frame if rotation is present.

6.10.2 Rotation

Consider a simple shear flow on a rotating table, with lab basis vectors $\boldsymbol{e}_{1,2,3}$ and rotating basis vectors $\boldsymbol{e}_{x,y,z}$.

If the rotation vector $\boldsymbol{\Omega}$ is along $\boldsymbol{e}_z$ then

$$
\begin{pmatrix} \boldsymbol{e}_x(t) \\ \boldsymbol{e}_y(t) \\ \boldsymbol{e}_z \end{pmatrix} = \begin{pmatrix} \cos\Omega t & \sin\Omega t & 0 \\ -\sin\Omega t & \cos\Omega t & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \boldsymbol{e}_1 \\ \boldsymbol{e}_2 \\ \boldsymbol{e}_3 \end{pmatrix}
$$

Note that the two frames coincide at $t = 0$. The flow has velocity $\boldsymbol{u} = \dot{\gamma}y\boldsymbol{e}_x +$ rotation. In the table frame, the rotation does not contribute to $\dot{\gamma}$ so we have

$$
\dot{\gamma} = \begin{pmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} = \dot{\gamma}(\boldsymbol{e}_x \boldsymbol{e}_y + \boldsymbol{e}_y \boldsymbol{e}_x)
$$

$$
\implies \tau = \int_{-\infty}^{t} G(t - t')\dot{\gamma}\, \mathrm{d}t' = \dot{\gamma} \int_{-\infty}^{t} G(t - t')\, \mathrm{d}t' = \eta_{\{x,y\}}\dot{\gamma}
$$

where for any linear viscoelastic fluid,

$$\eta_{\{x,y\}} = \int_0^\infty G(s)\,\mathrm{d}s$$

In the lab frame, we use the expressions for $\boldsymbol{e}_{x,y,z}$ in terms of $\boldsymbol{e}_{1,2,3}$ to get

$$\begin{aligned}
\dot{\boldsymbol{\gamma}} &= \dot{\gamma}(\boldsymbol{e}_x\boldsymbol{e}_y + \boldsymbol{e}_y\boldsymbol{e}_x) \\
&= \dot{\gamma}\begin{pmatrix} -\sin(2\Omega t) & \cos(2\Omega t) & 0 \\ \cos(2\Omega t) & \sin(2\Omega t) & 0 \\ 0 & 0 & 0 \end{pmatrix}
\end{aligned}$$

with respect to the $\boldsymbol{e}_{1,2,3}$ basis. The relevant deviatoric stress τ_{12} in the lab frame is then

$$\tau_{12} = \int_{-\infty}^t G(t-t')\dot{\gamma}\cos 2\Omega t'\,\mathrm{d}t'$$

$$\overset{s=t-t'}{=} \int_0^\infty G(s)\dot{\gamma}\cos 2\Omega(t-s)\,\mathrm{d}s$$

At $t=0$, both frames coincide so we expect the viscosity is the same in both frames at this time. We find

$$\tau_{12}\big|_{t=0} = \dot{\gamma}\int_0^\infty G(s)\cos 2\Omega s\,\mathrm{d}s = \eta_{\{1,2\}}\dot{\gamma}$$

where $\eta_{\{1,2\}} = \int_0^\infty G(s)\cos 2\Omega s\,\mathrm{d}s \neq \eta_{\{x,y\}}$. Hence we find the viscosity depends on the frame used to evaluate it. This is a fundamental problem with the LVF constitutive relaetionship. We can fix this by introducing *objective derivatives* for tensors. Note $G(s)$ has support when $s = \mathcal{O}(\lambda)$. Therefore in the integral for $\eta_{\{1,2\}}$, if $\Omega\lambda \ll 1$ then $\cos 2\Omega s \sim 1$ so the viscosities agree. Hence we can use the LVF model for flows with small velocity gradients or very short memory.

7 Objective Derivatives

7.1 Oldroyd's axioms

Newtonian fluid mechanics was developed in the 19th century. The theory of complex fluids was developed in the 20th century. In the 1950s, there was intense research to set up a rigorous mathematical framework for continuum mechanics. There are two different approaches:

- *Bottom up:* start with a microscopic model and from this form field equations via averaging. For example, polymers, statistical physics, ensemble avergaging gives field equations for density and velocity.

- *Top down:* axiomatic approach. Consider all terms that are admissible in a hydrodynamic equation based on symmetries or invariance principles. Fit the resulting free parameters to experimental data.

The game changer was Professor James Oldroyd who specified four axioms a constituve equation must satisfy. A constitutive relationship must be based on:

1. the relative motion of the neighbourhood of the (current) particle;

2. the history of the strain tensor associated with the particles;

3. physical constants defining the behaviour of the material (and obey its symmetries);

4. a *convective* coordinate system that is *embedded* in the material and is *deforming* with it, i.e. can only involve derivatives that do not depend on the frame where they are evaluated but instead need to be *intrinsic*.

Thus far, we have satisfied the first three axioms. Axiom 4 requires the introduce of 'new' tensor derivatives.

7.2 Upper-convected derivative

Consider two frames.

1. The 'lab' Cartesian frame with basis vectors $\boldsymbol{e}_i$ and coordinates x_i.

2. Curvilinear coordinates deforming with the flow (*covariant*) with basis vectors $\boldsymbol{g}_j$ and coordinates a_j.
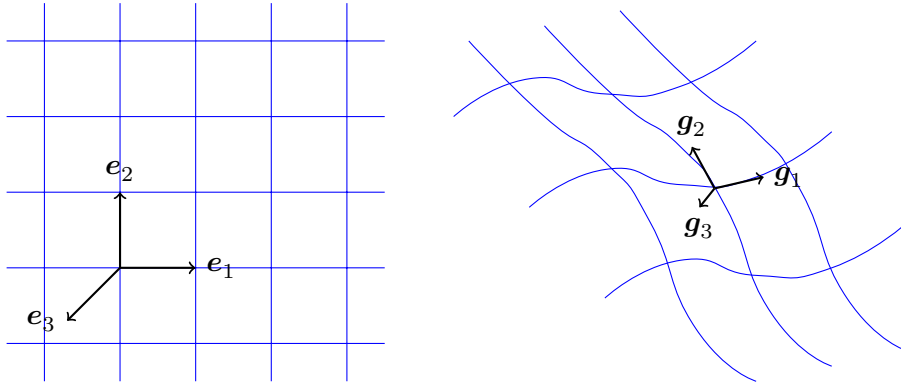
The covariant vectors $\boldsymbol{g}_i$ are *material vectors*, meaning they may change in length and orientation with the deforming material. The stress tensor in each frame is

$$\sigma = \begin{cases} \sigma_{mn}\boldsymbol{e}_m\boldsymbol{e}_n & \text{lab frame} \\ \hat{\sigma}_{ij}\boldsymbol{g}_i\boldsymbol{g}_j & \text{covariant frame} \end{cases}$$

A material point $\boldsymbol{x}$ is represented as $\boldsymbol{x} = x_j\boldsymbol{e}_j$ in the lap frame and $\boldsymbol{x} = a_j\boldsymbol{g}_j$ in the covariant frame. Hence by definition

$$\boldsymbol{g}_i = \frac{\partial \boldsymbol{x}}{\partial a_i} = \frac{\partial}{\partial a_i}\left[x_j\boldsymbol{e}_j\right] = \frac{\partial x_j}{\partial a_i}\boldsymbol{e}_j$$

Define the *deformation gradient tensor F* such that

$$F_{ij} = \frac{\partial x_j}{\partial a_i}$$

Then $\boldsymbol{g}_i = F_{ij}\boldsymbol{e}_j$. Using this frame transformation rule, we can form a tensor transformation rule for σ:

$$\begin{aligned} \sigma = \sigma_{mn}\boldsymbol{e}_m\boldsymbol{e}_n &= \hat{\sigma}_{ij}\boldsymbol{g}_i\boldsymbol{g}_j \\ &= \hat{\sigma}_{ij}F_{im}F_{jn}\boldsymbol{e}_m\boldsymbol{e}_n \\ \implies \sigma_{mn} &= \hat{\sigma}_{ij}F_{im}F_{jn} \end{aligned}$$

In tensor notation, this is equivalent to

$$\sigma = F^T \cdot \hat{\sigma} \cdot F \iff \hat{\sigma} = \left(F^T\right)^{-1} \cdot \sigma \cdot F^{-1}$$

Note that F is not unitary. We can now calculate the *intrinsic* time derivative of $\hat{\sigma}$:

$$\frac{\mathrm{D}}{\mathrm{D}t}\hat{\sigma} = \frac{\mathrm{D}}{\mathrm{D}t}\left[F^{-T} \cdot \sigma \cdot F^{-1}\right]$$

$$= \left(\frac{\mathrm{D}}{\mathrm{D}t}F^{-T}\right) \cdot \sigma \cdot F^{-1} + F^{-T} \cdot \left(\frac{\mathrm{D}}{\mathrm{D}t}\sigma\right) \cdot F^{-1} + F^{-T} \cdot \sigma \cdot \left(\frac{\mathrm{D}}{\mathrm{D}t}F^{-1}\right)$$

To elucidate this expression we wish to calculate the material derivative of F, using the fact $x_j = x_j(a_i, t)$. We find

$$\frac{\mathrm{D}}{\mathrm{D}t}F_{ij} = \frac{\mathrm{D}}{\mathrm{D}t}\frac{\partial x_j}{\partial a_i} = \frac{\partial}{\partial a_i}\frac{\mathrm{D}x_j}{\mathrm{D}t} = \frac{\partial u_j}{\partial a_i}$$

$$= \frac{\partial u_j}{\partial x_j}\frac{\partial x_k}{\partial a_i}$$

$$= F_{ik}\left(\nabla \boldsymbol{u}\right)_{kj}$$

$$\therefore \frac{\mathrm{D}}{\mathrm{D}t}F = F \cdot \nabla \boldsymbol{u}$$

Now we know $F \cdot F^{-1} = I$, hence

$$\frac{\mathrm{D}F}{\mathrm{D}t} \cdot F^{-1} + F \cdot \frac{\mathrm{D}F^{-1}}{\mathrm{D}t} = 0$$

$$\implies \frac{\mathrm{D}F^{-1}}{\mathrm{D}t} = -F^{-1} \cdot \frac{\mathrm{D}F}{\mathrm{D}t} \cdot F^{-1} = -(\nabla \boldsymbol{u}) \cdot F^{-1}$$

$$\frac{\mathrm{D}F^{-T}}{\mathrm{D}t} = -F^{-T} \cdot (\nabla u)^T$$

Collecting these results we have

$$\frac{\mathrm{D}\hat{\sigma}}{\mathrm{D}t} = F^{-T} \cdot \left[\frac{\mathrm{D}\sigma}{\mathrm{D}t} - (\nabla \boldsymbol{u})^T \cdot \sigma - \sigma \cdot (\nabla \boldsymbol{u})\right] \cdot F^{-1}$$

This is in the form of the frame transformation rule for tensors. Hence the intrinsic derivative of the stress tensor in the lab basis is

$$\overset{\triangledown}{\sigma} = \frac{\mathrm{D}\sigma}{\mathrm{D}t} - (\nabla \boldsymbol{u})^T \cdot \sigma - \sigma \cdot (\nabla \boldsymbol{u})$$

This is the *upper-convected derivative* (UCD) which is objective, i.e. independent of frame. It is the derivative measured intrinsically as it deforms with the flow. Note that

$$\overset{\triangledown}{I} = 0 - (\nabla \boldsymbol{u})^T \cdot I - I \cdot (\nabla \boldsymbol{u}) = -\dot{\gamma}$$

Other ways of defining objective derivatives include

- *Lower-convected derivative*

$$\overset{\triangle}{\sigma} = \frac{\mathrm{D}\sigma}{\mathrm{D}t} + (\nabla \boldsymbol{u})^T \cdot \sigma + \sigma \cdot (\nabla \boldsymbol{u})$$

- *Co-rotational derivative*

$$\frac{\mathcal{D}\sigma}{\mathcal{D}t} = \frac{\mathrm{D}\sigma}{\mathrm{D}t} + \frac{1}{2}\left(\boldsymbol{\omega} \cdot \sigma - \sigma \cdot \boldsymbol{\omega}\right)$$

where $\boldsymbol{\omega} = \nabla \boldsymbol{u} - (\nabla \boldsymbol{u})^T$.