

## 2 Geophysical Aspects of Non-Newtonian Fluid Mechanics

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### 2.1 Introduction

Non-Newtonian fluid mechanics is a vast subject that has several journals partly, or primarily, dedicated to its investigation (Journal of Non-Newtonian Fluid Mechanics, Rheologica Acta, Journal of Fluid Mechanics, Journal of Rheology, amongst others). It is an area of active research, both for industrial fluid problems and for applications elsewhere, notably geophysically motivated issues such as the flow of lava and ice, mud slides, snow avalanches and debris flows. The main motivation for this research activity is that, apart from some annoyingly common fluids such as air and water, virtually no fluid is actually Newtonian (that is, having a simple linear relation between stress and strain-rate characterized by a constant viscosity). Several textbooks are useful sources of information; for example, [1,2,3] are standard texts giving mathematical and engineering perspectives upon the subject. In these lecture notes, Aucey's chapter on rheology (Chap. 3) gives further introduction.

Non-Newtonian fluids arise in virtually every environment. Typical examples within our own bodies are blood and mucus. Other familiar examples are lava, snow, suspensions of clay, mud slurries, toothpaste, tomato ketchup, paints, molten rubber and emulsions. Chemical engineers, and engineers in general, are faced with the (often considerable) practical difficulties of modelling a variety of industrial processes involving the flow of some of these materials. Consequently, much theory has been developed with this in mind, and our aim in this review is to guide the reader through some of the developments and to indicate how and where this theory might be used in the geophysical contexts.

### 2.2 Microstructure and Macroscopic Fluid Phenomena

Most non-Newtonian fluids are characterized by an underlying microstructure that is primarily responsible for creating the macroscopic properties of the fluid. For example, a variety of non-Newtonian fluids are particulate suspensions – Newtonian solvents, such as water, that contain particles of another material. The microstructure that develops in such suspensions arises from particle–particle or particle–solvent interactions; these are often of electrostatic or chemical origin.

A common example of such a suspension is a slurry of kaolin (clay) in water. Kaolin particles roughly take the form of flat rectangular plates with different electrostatic charges on the faces and on the sides; their physical size is of the order of a micron. In static fluid, the plates stack together like a giant house of cards. This structure becomes so extensive that the electrostatic forces that hold the structure together engender a macroscopic effect, namely the microstructure is able to provide a certain amount of resistance to fluid flow [4].

Of course, the image of the kaolin structure within the slurry as a giant house of cards is a gross idealization. Undoubtedly, the kaolin forms an inhomogeneous, defective structure with a variety of length scales. Nevertheless, the important idea is that microstructure can lead to macroscopic observable effects on the flow of the fluid. For the kaolin slurry, we anticipate that microstructure adds to the resistance to flow provided the shearing (rate of deformation) is not too great. However, once the fluid is flowing and shearing over relatively long scales, the microstructure must disintegrate – the house of cards collapses. Thus, for greater shearing (larger rates of deformation), the fluid begins to flow more easily. This macroscopic, non-Newtonian effect of “shear thinning” is well documented and a key effect in suspension mechanics. The crudest model of the phenomenon is to make the fluid viscosity a decreasing function of the rate of strain. In this simple departure from the regular fluid behaviour, one then makes the shear stress a nonlinear function of the strain rate. This is an example of a “constitutive law”; we elaborate further on such laws soon, but first we continue with a brief discussion of other non-Newtonian effects.

If the concentration of kaolin is sufficiently high, the microstructure can provide so considerable a resistance to deformation that material does not flow at all until a certain amount of stress is exerted on the fluid. At smaller stresses, the fluid behaves like an elastic solid, and simply returns to its original state if the applied stress is removed. Above the critical stress, the “yield stress”, the material begins to flow. Materials exhibiting yield behaviour are said to behave plastically, and when they flow viscously after yield, the terminology viscoplastic is often used.

The kaolin–water slurry is what one might call a “pure” form of mud. But, when the mud is less pure, and contains numerous embedded particles, grains or boulders with widely varying sizes (as in most geophysical conditions), the clay particles still form microstructure, with the attendant macroscopic effects. Hence muds are a classic example of a geophysical viscoplastic fluid. But there are also other geophysical materials with microstructure. For example, snow flakes, through a process of partial melting and refreezing, act to form a static coherent structure; this is relevant when considering avalanches, see also Chap. 13. And lava has a microstructure of bubbles and silicate crystals suspended within a hot viscous solvent.

Shear thinning and yield stresses are common effects in particle suspensions, but they are not the only type of non-Newtonian behaviour we can encounter. Another type of behaviour arises in polymeric fluids. Here, the fluid is laced with high molecular weight deformable molecules (polymers), whose length can

be so long that the collective effect of the deformations of individual molecules affects the flow. Notably, because polymers coil and entangle themselves and their neighbours through weak molecular interactions (such as hydrogen bonding), they provide an effective elastic force that resists flow deformations which separate, straighten and stretch them. Moreover, because the forces produced by molecular rearrangements depend on their original orientations, polymeric fluids can also display significant memory dependence; that is, the fluid “remembers” the way in which it has been deformed. The macroscopic consequence is that the fluid can display highly elastic effects, such as the recoil of the fluid back into a container after it has begun to pour out of it.

Some of the effects of such “viscoelasticity” can be rather weird and surprising, and in all discussion of such fluids it is customary to mention a few examples: The Weissenberg effects [5] include die swell [6,7], wherein fluid emerges from a pipe and then undergoes a subsequent and sudden radial expansion downstream, and rod climbing, where the free surface of a rotating fluid rises up around the rod forcing it into motion (the surface of a Newtonian fluid would be depressed there). In the flow of a viscoelastic liquid down an open channel, the free surface bulges slightly to create a rounded fluid profile [8]. Viscoelastic flow past a bubble [9] leads to a distinct cusp at the rear stagnation point due to a long filament of highly stretched polymers in the bubble wake.

An important point that one should take from this discussion is that non-Newtonian fluid effects can be varied and unusual. As a result, the literature on non-Newtonian fluid mechanics contains many models of suspensions and polymeric fluids, each adding or encapsulating some observed effect. Unfortunately many of these models are designed with precisely one set of effects in mind and none adequately deal with the general non-Newtonian fluid. Consequently, because non-Newtonian effects all typically stem in some way from the underlying fluid microstructure, one should keep the microscopic physics in mind whilst negotiating one’s way through the minefield of rheological models to which we now give some introduction.

## 2.3 Governing Equations

To begin, we must first describe the continuum approximation that underlies the models to be discussed here. This continuum approximation assumes that the dimensions of the flow fields we are considering, with lengthscale  $L$ , are far greater than the lengthscale of the microstructure of the fluid  $l$ ; that is,  $L \gg l$ . Given this continuum hypothesis we can derive the governing equations for a fluid using conservation of mass and examining the rate of change of momentum within a volume of fluid with lengthscale  $L$ . If the fluid is incompressible, mass conservation yields

$$\nabla \cdot \mathbf{u} = 0, \quad (2.1)$$

where  $\mathbf{u}$  denotes the Eulerian velocity field (here we shall only consider incompressible fluids). Conservation of momentum leads us to

$$\varrho \frac{D\mathbf{u}}{Dt} = \nabla \cdot \boldsymbol{\sigma} + \mathbf{F}, \quad (2.2)$$

where the fluid density is  $\varrho$ , the convective derivative is  $D/Dt \equiv \partial/\partial t + \mathbf{u} \cdot \nabla$ , the stress tensor is  $\boldsymbol{\sigma} \equiv \{\sigma_{ij}\}$ , and  $\mathbf{F}$  denotes a body force, such as gravity. For incompressible fluids, the stress tensor is conveniently split into an isotropic piece  $-p\mathbf{I}$ , where  $p$  is the pressure field, and a remainder, here denoted by  $\boldsymbol{\tau} \equiv \{\tau_{ij}\}$ , called the deviatoric stress tensor. Thus,

$$\boldsymbol{\sigma} = -p\mathbf{I} + \boldsymbol{\tau} \quad \text{or} \quad \sigma_{ij} = -p\delta_{ij} + \tau_{ij}, \quad (2.3)$$

and the momentum equation becomes

$$\varrho \frac{D\mathbf{u}}{Dt} = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \mathbf{F}. \quad (2.4)$$

So far, apart from the continuum hypothesis, and for brevity and practicality assuming incompressibility, we have not made any statement about the fluid itself; mass conservation and the momentum equation are valid for all fluids. Thus the development so far parallels that of a Newtonian fluid, much as can be found in textbooks such as [10].

To produce a closed model, we must further specify how the deviatoric stress tensor  $\tau_{ij}$  is related to the properties of the fluid. Many non-Newtonian fluid models do this by relating the deviatoric stress to the rate-of-strain tensor,  $\dot{\gamma}_{ij}$ , here defined as

$$\dot{\gamma} = \nabla \mathbf{u} + (\nabla \mathbf{u})^T \quad \text{or} \quad \dot{\gamma}_{ij} = \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}; \quad (2.5)$$

where the superscript  $T$  denotes the transpose (some other authors use a minor variation with an extra factor of 1/2). Further variables are also sometimes included, such as the strain tensor  $\gamma_{ij}$  (which arises in linear elasticity), temperature, pressure, or particulate concentration. The relationship between  $\tau_{ij}$ ,  $\dot{\gamma}_{ij}$  and any other variables is the *constitutive relation* of the fluid, and closes the set of governing equations. This relation is the key ingredient to non-Newtonian fluid models and contains all of the fluid microphysics; unsurprisingly, the constitutive law can be extremely complicated. Indeed, there is considerable freedom in deciding how the fluid behaves due to changes in its deformation (the instantaneous strain, strain rates or strain history), or the behaviour due to its surroundings (such as temperature or pressure).

If the fluid is temperature-dependent and in a situation where the temperature can change, as is often the case for ice or lava flows, then we also require an energy equation. This equation describes, for example, how mechanical energy is converted by molecular friction into heat. Such frictional heating is often negligible in many fluid problems – after all we do not heat cups of coffee by stirring

them. But in ice flows, this effect can be important (see Chap. 11). Of much more importance in general fluid problems, however, is that a change in temperature can affect the fluid microstructure. This may give rise to magnitudes of variation in macroscopic material properties. Indeed, many fluids are Newtonian at fixed temperature, but have viscosities that are dramatically affected by temperature changes, as spreading golden syrup upon hot toast will demonstrate.

The energy equation is:

$$\rho c \frac{DT}{Dt} = \frac{1}{2} \tau_{ij} \dot{\gamma}_{ij} + \nabla \cdot (\mathcal{K} \nabla T) . \quad (2.6)$$

The parameters  $c$  and  $\mathcal{K}$  are the specific heat (at constant pressure or volume, as the fluid is incompressible) and conductivity. In deriving this equation we have assumed that the thermal expansion coefficient for the fluid is negligible, and we have ignored other energy sources or sinks, such as from plastic or elastic work, or from inelastic collisions between particles within the microstructure. The energy equation describes how the temperature field evolves in the fluid as a result of advection, diffusion and frictional heating. Such thermal evolution subsequently affects fluid microstructure and, thence, material properties. In turn, this modifies the fluid flow according to the constitutive law.

## 2.4 Constitutive Models

Newtonian fluids are characterized by an isotropic microstructure of passive spherical molecules that do not chemically interact with one another. The constitutive law is particularly simple: the deviatoric stress is linearly proportional to the rate of strain and the coefficient of proportionality is the viscosity,  $\mu$ . Thus

$$\tau_{ij} = \mu \dot{\gamma}_{ij} ,$$

and (2.2) reduces to the more familiar Navier–Stokes equation,

$$\rho \frac{D\mathbf{u}}{Dt} = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{F} .$$

For non-Newtonian fluids the constitutive relations can be much more complicated and must be built to reflect the macroscopic properties engendered by the fluid microstructure. There are several ways in which one goes about this construction; here we mention four different styles.

The first kind of approach is theoretical and “kinetic”: one assembles a model of the molecular anatomy of the fluid and then builds a kinetic theory for the fluid microstructure. Sometimes, this goes by way of an investigation of the flow around a single idealized model polymer, or emulsion droplet, and then the generation of the appropriate constitutive equation for a dilute suspension via an averaging procedure [11]. But other routes are also possible, including the representation of the fluid microstructure as a regular lattice or network of interacting elements [12]. These theories furnish a fluid model directly from the

input microscopic physics, and in an idealized world would be the most sensible approach. Unfortunately, such kinetic approaches have only recently become possible, and even then only for very simple fluids. Moreover, the mathematics behind them is often based upon physical approximations rather than asymptotic analysis. The problem is that it is currently technically impossible to build a kinetic theory for anything more than a very simple range of molecular models. For example, a popular model in visco-elasticity is a perfect network of identical elastic rods. But real fluids never conform to the idealizations necessary in order to fabricate kinetic theories, and even the simplest of such theories can lead to constitutive laws with very convoluted forms. Nevertheless, much progress has been made in the recent non-Newtonian fluid literature in this direction.

A second style of approach is purely phenomenological: one simply writes down a convenient model equation that represents how one imagines the fluid microstructure to affect the flow. Historically, this type of approach was the first used in non-Newtonian fluid mechanics. For example, Maxwell's model of a viscoelastic fluid was largely phenomenological – the stresses have a “fading memory” of the strain rates, which models the relaxation of the fluid to applied deformation at a molecular level.

The third approach was taken somewhat after the first phenomenological models and is largely an attempt to improve on them. The phenomenological theories provided a set of simple constitutive relations that at times did not possess some of the symmetries of the fluid. For example, the original Maxwell model was not “objective” when written in three dimensions, meaning that it took different forms in different frames of reference (see later). The third approach was therefore to write down the simplest kinds of constitutive models that possessed the same symmetries as the fluid. Thus Oldroyd wrote down a general constitutive model for a linear visco-elastic fluid model. This “Oldroyd-8” model contains a set of free parameters and has been claimed to work well in several situations. Moreover, several kinetic theories have also eventually led to the same kinds of models.

The difficulty in proceeding theoretically to furnish the constitutive law has led to a very popular fourth approach which is practical, but empirical. One performs various experiments upon the fluid using, for example, a viscometer, and then postulates a plausible stress strain-rate relation. Experiments for non-Newtonian fluids are not necessarily easy to perform [6] and a considerable amount of effort is sometimes required to neatly design experiments that isolate a particular factor. This empirical approach focusses on the macroscopic behaviour of the fluid and to a large extent simply takes the fluid microstructure for granted. Needless to say, the empirical models that one derives in this way are dangerous in that they are derived for specific experimental conditions and are not necessarily suitable once one changes those conditions. However, given some non-Newtonian fluid with a complicated and possibly unknown microstructure, the empirical approach is often the most expedient way forward.

This discussion should illustrate to the reader how non-Newtonian fluid mechanics has a certain schizophrenic aspect to it. On the one hand, the theory is

mathematically complicated and furnishes unwieldy constitutive laws. And on the other, there is a pragmatic approach that provides workable, but potentially unreliable, models. Below we give some examples of the forbidden fruit of the marriage of the two.....

## 2.5 Generalized Newtonian Models

Generalized Newtonian fluid models assume a fairly simple constitutive relation in which one modifies the linear relationship between the stresses and the strain rates by making the constant of proportionality, the viscosity, a prescribed function of strain-rate, temperature or particulate concentration. Thus,

$$\tau_{ij} = \mu(\dot{\gamma}, T, \phi) \dot{\gamma}_{ij} \quad (\text{Generalized Newtonian model}), \quad (2.7)$$

where we use  $\tau$  and  $\dot{\gamma}$  to represent the second invariants of the stress and strain rate,

$$\tau = \sqrt{\tau_{ij}\tau_{ij}/2}, \quad \dot{\gamma} = \sqrt{\dot{\gamma}_{ij}\dot{\gamma}_{ij}/2}; \quad (2.8)$$

( $\dot{\gamma}$  can be thought of as a measure of the magnitude of the deformation rate) and  $\phi$  is the particle concentration.

### 2.5.1 Power-law Fluids and the Herschel–Bulkley Model

A popular example of this kind of model is the power law fluid:

$$\mu(\dot{\gamma}) = K\dot{\gamma}^{n-1} \quad (\text{power law model}). \quad (2.9)$$

This viscosity function has two parameters, the consistency  $K$  and the index  $n$ . If  $n = 1$  we revert to Newtonian behaviour and the consistency is just the viscosity. If  $n < 1$ , the effective viscosity decreases with the amount of deformation. Thus this models the disintegration of fluid structure under shear, the shear thinning effect mentioned earlier.

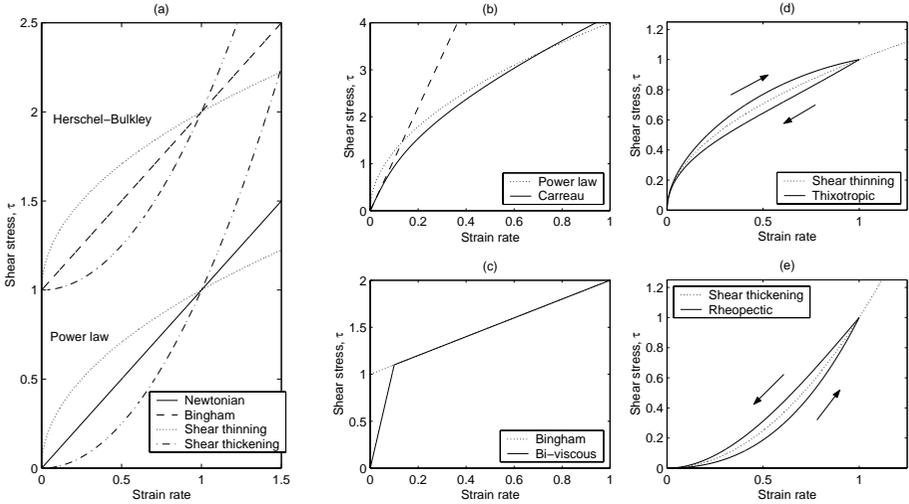
Conversely, if  $n > 1$ , the viscosity increases with the amount of shearing, which implies that the fluid microstructure is build up by the fluid motion. This kind of effect can occur if the molecules of the microstructure can bind together on contact; during increasing flow these molecules can come into contact more regularly and thus larger structures are created. Examples of such “shear thickening” materials are corn flour (which is used to thicken soup) and highly concentrated suspensions. The latter show shear thickening due to dilatancy [3]: at low shear rates the particles are closely packed together and a small amount of fluid lubricates the flow of particles. But, at higher shear rates, the close packing is disrupted and the material expands (dilates) and there is no longer enough fluid to lubricate particle–particle interactions. The resistance to flow then increases substantially.

The empirical power law model is a useful fit to the observed data, and can often provide good quantitative results over many decades of the shear rate.

However, it does not capture the effects of yield stress. Probably the most popular model that incorporates both shear thinning or thickening and a yield stress is the Herschel–Bulkley model [13]:

$$\begin{aligned} \tau_{ij} &= (K\dot{\gamma}^{n-1} + \tau_p/\dot{\gamma}) \dot{\gamma}_{ij} & \text{for } \tau \geq \tau_p \\ \dot{\gamma}_{ij} &= 0 & \text{for } \tau < \tau_p \end{aligned} \quad (\text{Herschel – Bulkley model}). \tag{2.10}$$

The new parameter  $\tau_p$  that we introduce is the yield stress. This formula also contains an even simpler model, the Bingham fluid, which is given by (2.10) with  $n = 1$ . For this model, the fluid flows as a Newtonian fluid, with strain rate proportional to the difference between the applied and yield stresses, once it has yielded. With  $n \neq 1$ , the Herschel–Bulkley model allows also for shear thinning or thickening beyond yielding. Two recent review articles upon yield stress phenomena are [14] and [15] and the model often appears in geophysical models, as illustrated in several other chapters in this volume.



**Fig. 2.1.** Non-Newtonian fluid models. A sketch of the constitutive models for a variety of rheological models. In (a) we show the power-law and Herschel–Bulkley models. Three curves are shown in each case, displaying shear-thinning and shear-thickening flow curves. The Bingham fluid and a Newtonian fluid are also shown. In panel (b) we display the Carreau model,  $\mu(\dot{\gamma}) = \mu_\infty + (\mu_0 - \mu_\infty) / [1 + (\lambda\dot{\gamma})^2]^{(1-n)/2}$  ( $\mu_0$ ,  $\mu_\infty$ ,  $\lambda$  and  $n$  are constants), which regularizes the infinite viscosity of the shear-thinning power-law fluid at zero strain rate. In (c) we show the bi-viscous regularization of the Bingham model, which allows flow for all strain rates. Panels (d) and (e) show thixotropic and rheopectic hysteresis curves. The scales are arbitrary

### 2.5.2 Variants and Deviants

There are many other empirical equations that provide stress-strain-rate relations within the generalized Newtonian framework, although the power law, Bingham and Herschel–Bulkley models are those most widely used; an illustration showing these models is in Fig. 2.1. However, this is not to say that they are uniformly accepted. Indeed, there is much discussion in the recent literature over whether these models are physically plausible. For example, the shear-thinning power law fluid predicts an infinite viscosity at zero strain rate. Even the concept of a yield stress has received much recent criticism, with evidence presented to suggest that most materials weakly yield or creep near zero strain rate [15]. Moreover, from a mathematical perspective, the discontinuous surface defined by the yield condition,  $\tau = \tau_p$ , introduces several undesirable features into the non-Newtonian fluid model, mainly because this surface is difficult to track accurately. Such criticisms have fuelled the introduction of further models that go some way to avoid the problems (see [3] p. 14, and [1]). For example, the Carreau model regularizes the infinite viscosity of the shear-thinning power-law fluid (see Fig. 2.1). And various regularizations of the Herschel–Bulkley or Bingham fluid modify the constitutive law so that, for  $\dot{\gamma} \rightarrow 0$ , the stress abruptly decreases to zero in the manner of a Newtonian fluid with a large viscosity. The latter regularizations allows flow to occur even at very low strain rates and are particularly useful for numerical work, [16,17,18]. A popular, although not necessarily optimal, regularization is to adopt a biviscous model, as shown in Fig. 2.1.

Many geophysical materials such as muds [19,20], debris flows and snow avalanches (see Chaps. 13 and 21) display behaviour that can be crudely captured by the Herschel–Bulkley model. However, there are probably many other properties of these flows that cannot [21]. Nevertheless, at the very least, the Herschel–Bulkley model can be used as the starting point for more elaborate models. This model has also been used for lavas (see Chap. 7). Here, the microstructure is provided by a combination of bubbles and crystals. Bubbles deform with the fluid motion; numerical computations with bubbly viscous fluids suggest that shear thinning can result [22]. Crystals, however, may have the opposite effect [23]: crystallization can be induced by the shearing motion of the fluid and so microstructure can be build up in a shear thickening fashion. Both effects may compete in lava, and which dominates depends on the ambient conditions.

### 2.5.3 Temperature Dependence

Many materials have strongly temperature-dependent microstructure. For generalized Newtonian fluids, the most common way of accounting for this dependence is to make the viscosity a function of temperature. A popular choice is an exponential, Arrhenius, dependence:

$$\mu(T) = \mu_* \exp(Q/RT) \quad (2.11)$$

where  $\mu_*$  is the viscosity value evaluated at some reference temperature,  $Q$  is the activation energy and  $R$  is the universal gas constant. Sometimes it is more convenient to use the approximation,

$$\mu(T) = \mu_* \exp[-\tilde{G}(T - T_a)], \quad (2.12)$$

where  $T_a$  and  $\tilde{G}$  are two more prescribed constants. Provided the temperature variation is relatively small, (2.12) can be considered as an approximation to (2.11); in some other contexts, this is referred to as the Frank–Kamenetski approximation. Exponential forms for the temperature dependence are commonly used for lavas [23,24,25,26,27], laboratory materials used to model magma and lava (such as wax, paraffin and corn syrup [28,29]), muds [30,31,32], and ice sheets [33].

Some fluids display both strong temperature dependence and other non-Newtonian effects, like shear thinning or yield behaviour. Lava and ice are two such materials. Within those subjects there have been attempts to generate empirical models incorporating all these features. Typically, they proceed by simply combining the earlier models. For example, one particular model that has found a niche of geophysical importance is Glen’s Law [34,35] for the flow of ice. It has the stress-strain-rate relation,

$$\mu(\dot{\gamma}, T) = \exp(Q/nRT)\dot{\gamma}^{(n-1)/n}, \quad n \sim 3, \quad (2.13)$$

and combines an Arrhenius temperature dependence with shear thinning. Typically the constitutive law is written in terms of the second invariant of the stress, rather than the strain rate, for reasons of algebraic ease in subsequent analysis. However, despite the wide usage of this law, there is significant disagreement between measurements taken in various laboratory experiments and from actual ice flows [36]. Part of the reason for this disagreement seems to be that ice relaxes under stress only over long times, and this relaxation has not been correctly taken into account in most measurements.

#### 2.5.4 Concentration Dependence

Another issue that often arises in fluid suspensions is how the microstructural effects depend upon the particle concentration,  $\phi$ . For Newtonian fluids, the Einstein relation was deduced to give the viscosity correction due to a dilute suspension of rigid spheres within a solvent of viscosity  $\mu_0$ :

$$\mu = \mu_0 \left( 1 + \frac{5}{2}\phi \right). \quad (2.14)$$

Strictly speaking, this model is only suitable if the suspension is very dilute. A simple resummation of (2.14) that attempts to extend the formula to much larger concentrations is the Einstein–Roscoe relation:

$$\mu = \mu_0 \left( 1 - \frac{\phi}{\phi_m} \right)^{-\alpha}. \quad (2.15)$$

The quantity  $\phi_m$  is a maximum packing fraction beyond which the suspension cannot flow; for a suspension of solid spheres,  $\phi_m \approx 0.68$ , but this quantity depends on the shape of the particles and how they organize themselves into a lattice structure. Experiments with concentrated non-colloidal suspensions [37] suggest that a good empirical fit is achieved if  $\alpha \approx 1.82$ . Other related models are reviewed in [38]. Similar approximations have been developed for lava, where one argues that the role of the suspended particles is played by silicate crystals [39], and in temperate ice (a binary mixture of ice and water at the melting temperature), where the concentration does not refer to particles at all, but to the water content [40].

Particle concentration also affects the yield stress in viscoplastic fluids [14], and so we need another formula for  $\tau_p(\phi)$  in the constitutive law. In geophysical contexts, the combined effect of concentration dependence on viscosity and yield stress may be important for lava (because crystallization occurs when the temperature falls) and for some debris flows.

Given that the fluid properties depend on particle concentration, one should also add an equation that determines  $\phi$ . In some situations, it may be possible to treat the concentration as though it were homogeneous; then  $\phi$  is simply a parameter. However, the origin of many effects observed in suspensions can be traced to the appearance of an inhomogeneous particle distribution. A notable example that plagues chemical engineers is wall slip. Many rheometers operate by creating a shear flow inside the fluid by rotating the walls containing the material. Often it is observed that high shear layers build up near these walls in which the particle concentration is depleted. Because the fluid is then relatively dilute in these region, and they are frequently extremely thin, they act like lubricating “slip” layers. As a result, the direct measurements taken with the instrument can be in error.

Another example that may be of geophysical relevance is viscous resuspension. The observation here is that particles in a shearing suspension tend to migrate away from regions with relatively large shear. This migration provides an uplift in flows over plates that can oppose and even dominate the natural tendency to sediment [41].

To deal with concentration variations, we need a conservation equation for  $\phi$ . One relevant to viscous resuspension is [42] :

$$\frac{D\phi}{Dt} + \nabla \cdot [J_c + J_\mu] = 0 \quad (2.16)$$

$$J_c = -K_c a^2 \phi \nabla(\phi \dot{\gamma}), \quad J_\mu = -K_\mu \dot{\gamma} \phi^2 \frac{a^2}{\mu} \frac{d\mu}{d\phi} \nabla \phi. \quad (2.17)$$

Here  $J_c$  and  $J_\mu$  are the fluxes due to particle collisions and spatially varying viscosity; the particular forms quoted are given by heuristic arguments in [42]. The parameters  $K_c$  and  $K_\mu$  are constants determined experimentally and  $a$  is the particle radius.

In lava, particle diffusion and migration may be unimportant for silicate crystals. However, crystals form when the temperature decreases, and so one

should add sources and sinks associated with the phase change of solidification. Moreover, as in ice, the crystal structure may form anisotropically and with a broad distribution of sizes. The particle concentration  $\phi$  in lava could equally well be considered to be the concentration of bubbles or dissolved volatiles. We mentioned earlier the effect of bubbles, but volatiles add chemical effects that can also modify microstructure (for example,  $\text{OH}^-$  ions are observed to inhibit polymerization of silicon–oxygen bonds). Furthermore, as temperature and pressure changes, the bubble and volatile content can also change, with one being converted to the other. Overall, this makes the modelling of lava an extremely challenging problem.

### 2.5.5 Hysteresis

There are complicating issues that the generalized Newtonian models do not capture. One often overlooked issue is hysteresis. As described above, for a static viscoplastic material there is a microstructure that prevents flow until the yield stress is exceeded. Once flowing the structure is gradually broken down with increasing shear, and this gradual attrition of the microstructure leads to non-linear stress strain-rate behaviour. The reverse situation, in which the strain-rate is decreased until the structure reforms, is conceptually identical. However, there is no pressing reason why structure should reform in the same way that it disintegrates; in practice some hysteresis occurs. As a result the stress-strain-rate relation is not identical when the same material is measured with increasing or decreasing strain-rates. That is, the “up-curves” and “down-curves” on the  $\dot{\gamma}$ - $\tau$  plane are different.

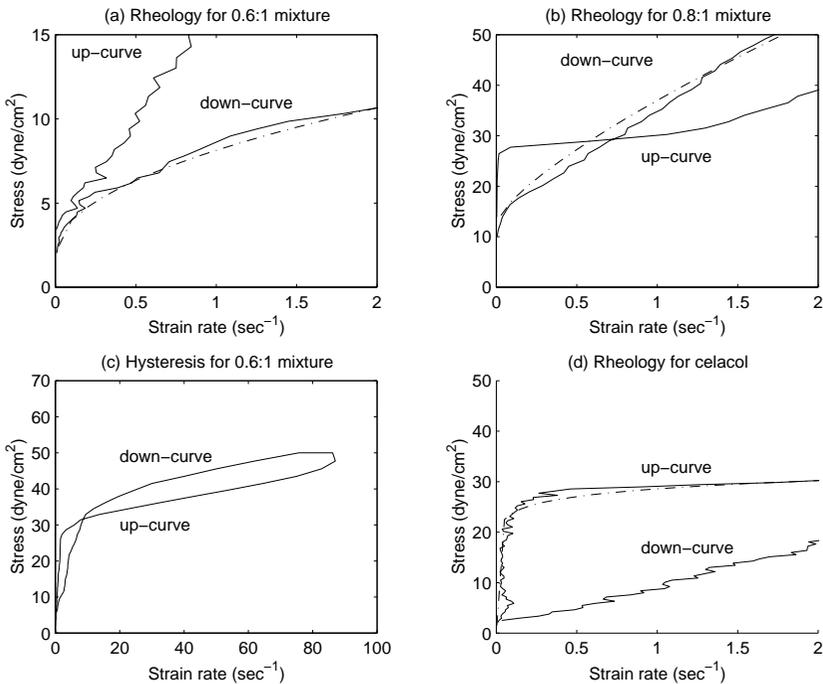
The most common types of hysteretic curves are illustrated in the final two panels of Fig. 2.1. The “thixotropic” fluid is shear thinning, and microstructure disintegrates due to the flow of the fluid. Thus the viscosity decreases during the experiment. The “rheopectic” fluid is shear thickening and structure builds up during the experiment. Both thixotropic and rheopectic behaviour have been observed in lavas [23]; thixotropy may be associated with the effects of bubbles, whereas shear-induced crystallization may be responsible for the rheopecty.

We illustrate hysteresis with some rheological measurements for a kaolin–water slurry and a celacol (Methyl–Cellulose) solution. The data is taken with a TI Instruments CSL 500 controlled-stress, cone-and-plate rheometer (6 cm, 2 degree measurement geometry). The results are shown in Fig. 2.2; this also shows the Herschel–Bulkley models that were used to fit the data. Hysteresis is certainly evident for the kaolin slurry. There are also some sharp changes in the up-curves that are possibly indicative of wall slip in the cone and plate device. The extreme example of celacol shows a material that behaves viscoplastically at first, but the destruction of the microstructure is permanent, and on decreasing the applied stress the material behaves viscously.

Another form of hysteresis occurs if the yield strength is itself time dependent, with a distinct gelation timescale. In this case, the structure that creates the yield strength takes time to form. Thus the material may have different yield strengths dependent upon when we choose to disturb it or bring it to rest [43].

**Table 2.1.** The properties of the experimental materials; the ratios are kaolin:water on a weight basis. Also shown are the parameters of the Herschel–Bulkley model from down-curves of stress sweeps with virgin material (see Fig. 2.2) using 6 cm 2 degree plate with CSL 500 Carrimed. †The Celacol data is taken from the up-curve

Material	0.6:1	0.8:1	1:1	1.2:1	Syrup	Celacol <sup>†</sup>
Density $\rho$ (g/cm <sup>3</sup> )	1.1	1.2	1.33	1.47	1.0	1.0
Yield stress $\tau_p$ (dyne/cm <sup>2</sup> )	20	130.0	500.0	1320.0	0.0	0.0
Consistency $K$ (units)	61	240	408	946	690	28.5
Index $n$	0.5	0.75	0.54	0.42	1.0	0.08



**Fig. 2.2.** The rheological data collected using a controlled stress sweep. Panels (a) and (b) show the stress strain-rate curves for the 0.6:1 and 0.8:1 kaolin–water mixtures. The up- and down-curves relate to whether the data was collected whilst the applied stress was increasing or decreasing; the dot-dash lines show the Herschel–Bulkley fit using the parameters of Table 2.1. Panel (c) shows the 0.6:1 data over a substantially extended range of strain-rates. The rheology of the celacol solution is shown in panel (d)

## 2.6 Viscoelasticity

Under some circumstances a material will exhibit both elastic and viscous behaviour; in response to some applied shear many materials show initially viscous behaviour and then ‘relax’ to elastic behaviour. The generalized Newtonian fluid model does not incorporate any elastic effects whatsoever, and so is inappropriate for such flows. Instead, it is usually necessary to introduce the strains as well as strain rates into the constitutive law. This is apparent from the form the constitutive law must take in the extreme limits: an incompressible linear elastic material has the stress is proportional to the strain, whereas a Newtonian fluid has the stress proportional to the rate of strain. Thus, for a general viscoelastic fluid, the constitutive law takes the form of an evolution equation.

The appearance of time evolution terms in the rheology relation reflects the relaxational character of the fluid stresses, and leads to the notion of a characteristic relaxation timescale. Many rheological measurement devices for viscoelastic fluids are designed with this in mind. One standard experiment is to apply instantaneously a shear at the surface of a sample material. If the material is linearly elastic the resulting stress is zero before the application of the shear, and constant immediately afterwards. On the other hand, if the material is a Newtonian fluid, the stress is infinite at the instant the stress is applied, but thereafter is zero. Thus elastic and viscous responses are markedly different, and many real materials have elements of both types of response. A viscoelastic material will have an initially large stress due to the viscous component, but the stress then decreases over the relaxation time to a constant value arising due to the intrinsic elasticity.

If we assume that the relation between the deviatoric stress and the strain rates is purely linear, then a general constitutive law can be stated:

$$\tau_{ij} = \int_{-\infty}^t G(t - \tau) \dot{\gamma}_{ij}(\tau) d\tau. \quad (2.18)$$

Here,  $G(t)$  is called the relaxation function, and builds in the elastic and viscous behaviour. Implicitly, the shape of the function  $G(t)$  determines the characteristic relaxation timescale (or timescales if there are more than one).

The relaxation time is important because it characterizes whether viscoelasticity is likely to be important within an experimental or observational timescale. For example, we might consider the continents upon the earth’s surface as solid over a timescale based upon the human lifespan, but upon a geological timescale they could be considered as a viscous, or viscoelastic, fluid. Many fluids, particularly those in industrial situations containing polymers or emulsion droplets, exhibit both elastic and viscous responses on an experimental or observational timescale.

For a Newtonian fluid,  $G(t) = \mu\delta(t)$  and relaxation is immediate. For a linear elastic material,  $G(t) = \mu H(t)$ . If we denote the relaxation time by  $\lambda$  then the simplest viscoelastic model, the Maxwell model, has  $G(t) = \mu \exp(-t/\lambda)/\lambda$  and the integral relation above can be recast in the form of a differential constitutive

relation,

$$\tau_{ij} + \lambda \dot{\tau}_{ij} = \mu \dot{\gamma} . \quad (2.19)$$

Much can be achieved with this simple extension to the Newtonian constitutive model, and in many circumstances, particularly if one wishes to investigate whether viscoelasticity can be important, this linear theory suffices. Extensions to multiple relaxation times with a sequence of relaxation functions are also straightforward.

Unfortunately, the Maxwell model (2.19) has at least one major failing – it is not frame indifferent (objective). That is, if we change to a moving coordinate frame the equations also change. Since we are concerned with material behaviour this should not occur. One crude, effective and ad-hoc cure is to replace the time derivatives in (2.19) with more complicated operators that build in the convection, rotation and stretching of the fluid motion. These operators, called either Oldroyd or Jaumann derivatives, render the equations frame indifferent; in usual tensor notation, the Oldroyd (upper convected) derivative,  $\overset{\nabla}{\mathbf{b}}$ , for a tensor  $\mathbf{b}$  is

$$\overset{\nabla}{\mathbf{b}} = \frac{D\mathbf{b}}{Dt} - \mathbf{b} \cdot (\nabla \mathbf{u}) - (\nabla \mathbf{u})^T \cdot \mathbf{b} \quad \text{or} \quad \overset{\nabla}{b}_{ij} = \dot{b}_{ij} + u_k b_{ij,k} - u_{j,k} b_{ki} - u_{i,k} b_{kj} . \quad (2.20)$$

These derivatives involve the local fluid motion, and so substantially complicate the constitutive law, and therefore computations using them.

Although we introduce these derivatives as a mathematical device to improve the linear model, one can also obtain these derivatives by working with dilute suspensions and low Reynold's number hydrodynamics – the kinetic approach mentioned earlier. By studying the fluid motion around a single elastic sphere, emulsion droplet, or a dumbbell connected with an elastic spring, and then analyzing the force exerted by the droplet upon the fluid, one can construct constitutive relations. Rather pleasingly these also involve Oldroyd, or Jaumann, derivatives and so the apparently crude mathematical fix has some physical basis. Further details of this approach can be found in [44] or [1].

A popular, more refined version of the Maxwell model is the so-called Oldroyd-B model; a simplification of his Oldroyd-8 model. The Oldroyd-B model takes account of the stresses due to both the Newtonian solvent and the polymeric constituents:

$$\boldsymbol{\tau} = \boldsymbol{\tau}_s + \boldsymbol{\tau}_p . \quad (2.21)$$

The total viscosity  $\mu$  is also written as the sum of solvent and polymeric viscosities,  $\mu = \mu_s + \mu_p$ . Thus, if  $\eta = \mu_s/(\mu_s + \mu_p)$ , the stress is written as

$$\boldsymbol{\tau} = \mu[\eta \overset{\nabla}{\boldsymbol{\gamma}} + (1 - \eta)\mathbf{a}] . \quad (2.22)$$

The constitutive equation for the extra stress tensor  $\mathbf{a}$  takes the form,

$$\mathbf{a} + \lambda \overset{\nabla}{\mathbf{a}} = \overset{\nabla}{\boldsymbol{\gamma}} , \quad (2.23)$$

where  $\lambda$  is the polymer relaxation time. There are several problems with the Oldroyd-B model [45], which suggest that it should not be used indiscriminately

to model viscoelastic flows. On the other hand, this model gives a reasonable description for some flows of dilute polymeric suspensions in highly viscous solvents with a single characteristic relaxation time (“Boger fluids” – [46]), and has been used extensively in attempting to characterize and interpret fluid flows [47,48].

One might imagine that because viscoelasticity is commonly engendered by dissolved polymers, there are few geophysical fluids which behave in this fashion. In fact, somewhat surprisingly, lava has been observed to show some viscoelastic non-Newtonian effects. For example, the Weissenberg effect (rod climbing) was observed in some laboratory experiments, and upward bulges have been seen on lava flows on Mount Etna [23]. Also, prolonged time-dependent relaxational effects are seen in measurements of density, pressure and sound speed [49]; relaxation times range from seconds to weeks.

## 2.7 Concluding Remarks

In this chapter we have given a brief overview of some phenomena and rheological models of non-Newtonian fluid mechanics. However, this is a notoriously involved subject, mainly due to the wide range of often complex and sometimes unexpected behaviours that real fluids and fluid-like materials exhibit. We can only hope to scratch the surface of the subject here, provide references to allow the interested reader to delve further into the subject, and draw together the underlying theory required in later chapters.

It is also important to appreciate the limitations of the models we have described. Indeed, this subject is not like Newtonian fluid mechanics where the Navier–Stokes equation is uniformly accepted; there is still much debate over which constitutive models are appropriate for different materials, and this is particularly prevalent for viscoelastic fluids. The generalized Newtonian models that seem easiest to use are empirical, and the explanation for the experimentally observed behaviour is based upon heuristic microstructural arguments. However, the models are essentially curve fits to observed data that have a convenient mathematical form. Some of the viscoelastic models have a sounder physical foundation, but they are typically far more complicated and are often designed with a specific phenomenon in mind and fail to incorporate the behaviour one wishes to model. None the less, many models exist with a spectrum of degrees of sophistication that build in both physical behaviour and mathematical niceties.

Despite all of these efforts much remains to be understood for non-Newtonian flows in general. Later chapters on debris flows, ice, snow avalanches and lava highlight aspects of the behaviours we have discussed in this chapter: yield stress, shear thinning, temperature dependence and particle concentration dependence. These chapters also describe the current modelling difficulties that remain. For example, the Bingham and Herschel–Bulkley models have had some success for concentrated mud flows containing fine particles [50,51], but have been less successful for flows containing larger particles [21]. Debris flows (Chap. 21) incorporate a range of particle sizes, that at one extreme may be so significant that

we violate the continuum approximation. The detailed failure of the Herschel–Bulkley model in these cases is due to several effects. The model does not allow for fluid motion relative to solid debris, it does not incorporate energy dissipation for the solid boulders and grains interacting, or for the way that such large objects can slide or roll along the base of the flow. None the less for primarily shear-dominated flows of concentrated suspensions of fine particles, Bingham-like models can provide good predicative and quantitative information. Indeed, in a later chapter we shall adopt the Herschel–Bulkley model to analyse some isothermal viscoplastic lava flows.

Lastly, we have focussed exclusively on fluids in this chapter. Yet some geophysical materials ought probably not to be treated as fluids at all. For example, the bubbly magma that rises through the conduits within volcanos (see Chap. 8) is much closer to being a foam, and dry landslides and avalanches and some debris flows [52] are fully fledged granular media (see Chap. 4).

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