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CHAPTER 1

CLASSIFICATION OF NON-NEWTONIAN FLUIDS

1.1 GENERAL CONSIDERATIONS AND DEFINITIONS

(a) Viscosity of Newtonian Fluids

Consider a thin layer of fluid between two parallel planes a distance dy apart as in Fig. 1.

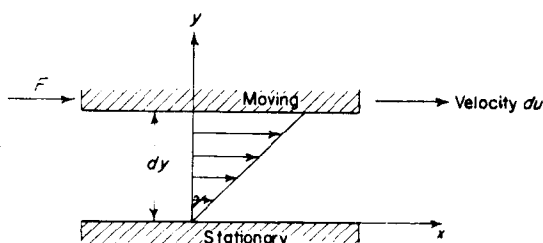


FIG. 1.

One plate is fixed and a shearing force F is applied to the other. When conditions are steady the force F will be balanced by an internal force in the fluid due to its viscosity. For a Newtonian fluid in laminar flow the shear stress is proportional to the velocity gradient, i.e.

$$F/A = \tau \propto du/dy$$

This equation may be written as

$$\tau = \mu du/dy = \mu \dot{\gamma} \quad [1.1.1]$$

where the constant of proportionality, μ , is called the Newtonian viscosity. It will be seen that μ is the tangential force per unit area exerted on layers of fluid a unit distance apart and having a unit velocity difference between them.

The Newtonian viscosity, μ , depends only on temperature and pressure and is independent of the rate of shear. The diagram relating shear stress and rate of shear for Newtonian fluids, the so-called 'flow curve', is therefore a straight line of slope μ as in Fig. 2, and the single constant, μ , completely characterizes the fluid.

Newtonian behaviour is exhibited by fluids in which the dissipation of viscous energy is due to the collision of comparatively small molecular species. All gases and liquids and solutions of low molecular weight come into this category. Notable exceptions are colloidal suspensions and polymeric solutions where the molecular species are large. These fluids show marked deviations from Newtonian behaviour.

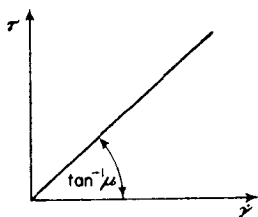


FIG. 2. Flow curve of a Newtonian fluid.

(b) Non-Newtonian fluids

Non-Newtonian fluids are those for which the flow curve is not linear, i.e. the 'viscosity' of a non-Newtonian fluid is not constant at a given temperature and pressure but depends on other factors such as the rate of shear in the fluid, the apparatus in which the fluid is contained or even on the previous history of the fluid.

These real fluids for which the flow curve is not linear may be classified into three broad types:

- (1) fluids for which the rate of shear at any point is some function of the shearing stress at that point and depends on nothing else;
- (2) more complex systems for which the relation between shear stress and shear rate depends on the time the fluid has been sheared or on its previous history;
- (3) systems which have characteristics of both solids and fluids and exhibit partial elastic recovery after deformation, the so-called viscoelastic fluids.

These three classes of fluids will now be treated in order.

1.2 TIME-INDEPENDENT NON-NEWTONIAN FLUIDS

Fluids of the first type whose properties are independent of time may be described by a rheological equation of the form

$$\dot{\gamma} = f(\tau) \quad [1.2.1]$$

This equation implies that the rate of shear at any point in the fluid is a

simple function of the shear stress at that point. Such fluids may be termed *non-Newtonian viscous fluids*.

These fluids may conveniently be subdivided into three distinct types depending on the nature of the function in Eqn. [1.2.1]. These types are

- (1) Bingham plastics
- (2) pseudoplastic fluids
- (3) dilatant fluids

and typical flow curves for these three fluids are shown in Fig. 3 and compared with the linear relation typical of Newtonian fluids.

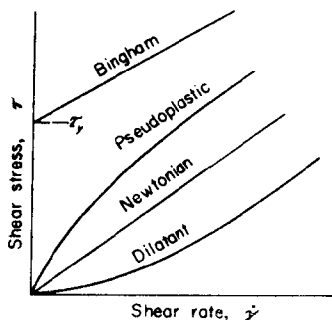


FIG. 3. Flow curves for various types of time-independent non-Newtonian fluids.

(a) Bingham plastics

A Bingham plastic⁽¹⁾ is characterized by a flow curve which is a straight line having an intercept τ_y on the shear-stress axis. The yield stress, τ_y , is the stress which must be exceeded before flow starts. The rheological equation for a Bingham plastic may be written

$$\tau - \tau_y = \mu_p \dot{\gamma} ; \tau > \tau_y \quad [1.2.2]$$

where μ_p , the plastic viscosity or coefficient of rigidity, is the slope of the flow curve.

The concept of an idealized Bingham plastic is very convenient in practice because many real fluids closely approximate this type of behaviour. Common examples are slurries, drilling muds, oil paints, toothpaste and sewage sludges. The explanation of Bingham plastic behaviour is that the fluid at rest contains a three-dimensional structure of sufficient rigidity to resist any stress less than the yield stress, τ_y . If this stress is exceeded the structure completely disintegrates and the system behaves as a Newtonian fluid under a shear stress $\tau - \tau_y$. When the shear stress falls below τ_y the structure is reformed.

(b) Pseudoplastic fluids

Pseudoplastic fluids show no yield value and the typical flow curve for these materials indicates that the ratio of shear stress to the rate of shear, which may be termed the apparent viscosity, μ_a , falls progressively with shear rate and the flow curve becomes linear only at very high rates of shear. This limiting slope is known as the viscosity at infinite shear and is designated μ_∞ .

The logarithmic plot of shear stress and rate of shear for these materials is often found to be linear with a slope between zero and unity. As a result, an empirical functional relation known as the *power law* is widely used to characterize fluids of this type. This relation, which was originally proposed by Ostwald⁽²⁾ and has since been fully described by Reiner,⁽³⁾ may be written as

$$\tau = k \dot{\gamma}^n \quad [1.2.3]$$

where k and n are constants ($n < 1$) for the particular fluid: k is a measure of the consistency of the fluid, the higher k the more viscous the fluid; n is a measure of the degree of non-Newtonian behaviour, and the greater the departure from unity the more pronounced are the non-Newtonian properties of the fluid. It is important to remember that although n is nearly constant in many cases over wide ranges of shear rate it is not a true constant for real fluids over all possible ranges of shear. This is not a serious drawback in engineering applications because all that is needed is a rheological equation which describes the fluid over the particular range of shear rate encountered in the particular problem. Over such a range n may often be regarded as constant.

It should be noted here that the dimensions of k depend on the index n and this fact has led to many objections to the use of the power law, e.g. by Reiner⁽³⁾. In most engineering applications these objections are not serious.

The apparent viscosity, μ_a , for a power law fluid may be expressed in terms of n since

$$\begin{aligned} \mu_a &= \tau / \dot{\gamma} \\ \text{i.e. } \mu_a &= k \dot{\gamma}^{n-1} \end{aligned} \quad [1.2.4]$$

and since $n < 1$ for pseudoplastics the apparent viscosity decreases as the rate of shear increases. This type of behaviour is characteristic of suspensions of asymmetric particles or solutions of high polymers such as cellulose derivatives. This suggests that the physical interpretation of this phenomenon is probably that with increasing rates of shear the asymmetric particles or molecules are progressively aligned. Instead of the random intermingled

state which exists when the fluid is at rest the major axes are brought into line with the direction of flow. The apparent viscosity continues to decrease with increasing rate of shear until no further alignment along the streamlines is possible and the flow curve then becomes linear.

Pseudoplastic fluids have been defined as time-independent fluids and this implies that the alignment of molecules suggested above takes place instantaneously as the rate of shear is increased or, at any rate, so quickly that the time effect cannot be detected using ordinary viscometric techniques.

Other empirical equations which have been used to describe pseudoplastic behaviour are

Prandtl	$\tau = A \sin^{-1}(\dot{\gamma}/C)$	
Eyring	$\tau = \dot{\gamma}/B + C \sin(\tau/A)$	
Powell-Eyring	$\tau = A \dot{\gamma} + B \sinh^{-1}(C\dot{\gamma})$	
Williamson	$\tau = A\dot{\gamma}/(B + \dot{\gamma}) + \mu_{\infty}\dot{\gamma}$	[1.2.5]

In these equations A , B and C are constants which are typical of the particular fluid. These equations are considerably more difficult to use than the power law and usually do not offer any compensating advantages.

(c) Dilatant fluids

Dilatant fluids are similar to pseudoplastics in that they show no yield stress but the apparent viscosity for these materials *increases* with increasing rates of shear. The power law equation is again often applicable but in this case the index n is greater than unity.

This type of behaviour was originally found in suspensions of solids at high solids content by Osborne Reynolds. He suggested that when these concentrated suspensions are at rest the voidage is at a minimum and the liquid is only sufficient to fill these voids. When these materials are sheared at *low* rates the liquid lubricates the motion of one particle past another and the stresses are consequently small. At higher rates of shear the dense packing of the particles is broken up and the material expands or 'dilates' slightly and the voidage increases. There is now insufficient liquid in the new structure to lubricate the flow of the particles past each other and the applied stresses have to be much greater. The formation of this structure causes the apparent viscosity to increase rapidly with increasing rates of shear.

The term 'dilatant' has since come to be used for all fluids which exhibit the property of increasing apparent viscosity with increasing rates of shear. Many of these, such as starch pastes, are not true suspensions and do not dilate on shearing in the normal sense of the word. The above explanation

therefore does not apply but nevertheless they are commonly referred to as 'dilatant' fluids.

In the process industries dilatant fluids are much less common than pseudo-plastic fluids but when the power law is applicable the treatment of both types is much the same.

1.3 TIME-DEPENDENT NON-NEWTONIAN FLUIDS

Many real fluids cannot be described by a simple rheological equation such as Eqn. [1.2.1] which applies to fluids for which the relation between shear stress and shear rate is independent of time. The apparent viscosity of more complex fluids depends not only on the rate of shear but also on the time the shear has been applied. These fluids may be subdivided into two classes:

(a) thixotropic fluids

(b) rheopectic fluids

according as the shear stress decreases or increases with time when the fluid is sheared at a constant rate.

(a) Thixotropic fluids—breakdown of structure by shear

Thixotropic materials are those whose consistency depends on the duration of shear as well as on the rate of shear.

If a thixotropic material is sheared at a constant rate after a period of rest, the structure will be progressively broken down and the apparent viscosity will decrease with time. The rate of breakdown of structure during shearing at a given rate will depend on the number of linkages available for breaking and must therefore decrease with time. (This could be compared with the rate of a first-order chemical reaction.) The simultaneous rate of reformation of structure will increase with time as the number of possible new structural linkages increases. Eventually a state of dynamic equilibrium is reached when the rate of build-up of structure equals the rate of breakdown. This equilibrium position depends on the rate of shear and moves towards greater breakdown at increasing rates of shear.

As an example we could consider the material confined in a cylindrical viscometer (see Chapter 6). After the material has been resting for a long time one of the cylinders is rotated at a constant speed. The torque on the other cylinder would then decrease with time as shown in Fig. 4. The rate of decrease and the final torque would both depend on the speed, i.e. on the rate of shear.

Thixotropy is a reversible process and after resting the structure of the material builds up again gradually. If the flow curve of a thixotropic material is determined immediately after shearing and after it has rested for varying times after shearing the result is as in Fig. 5.

This type of behaviour leads to a kind of hysteresis loop on the curve of shear-stress plotted against rate of shear if the curve is plotted first for the rate of shear increasing at a constant rate and then for the rate of shear decreasing at a constant rate. This is illustrated in Fig. 6, where the curves A and B are drawn for fluids of the Newtonian and pseudoplastic types which exhibit thixotropy.

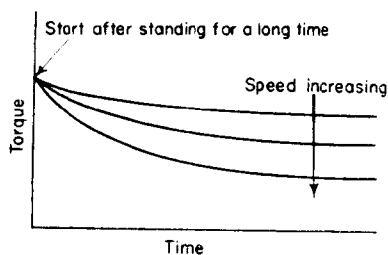


FIG. 4.

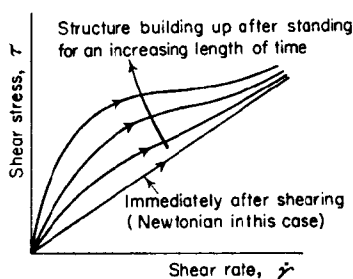


FIG. 5.

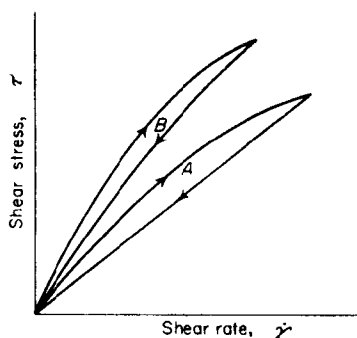


FIG. 6. Hysteresis loops for thixotropic fluids.

Flow curves of increasing height can be obtained by applying shear for increasing lengths of time before making the return path. A single curve can

be obtained by continuing the shearing process to equilibrium before returning.

The term 'false-body' is frequently encountered in discussions on thixotropy. This was introduced by Pryce-Jones⁽⁴⁾ to distinguish types of thixotropic behaviour of Bingham plastics. True thixotropic materials break down completely under the influence of high stresses and behave like true liquids even after the stress has been removed, until such time as the structure has reformed. False-bodied materials, on the other hand, do not lose their solid properties entirely and can still exhibit a yield value even though this might be diminished. The original yield value is only regained after resting for a long time.

The hysteresis loop on the flow curve would take the form of Fig. 7 for these two materials.

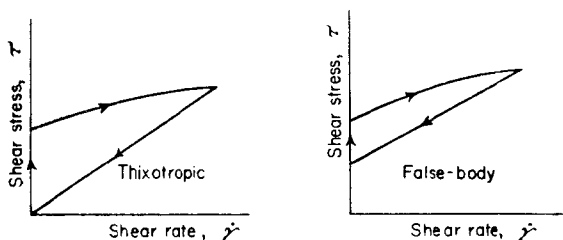


FIG. 7.

This behaviour can be illustrated by the following experiment. Consider a liquid in a vessel with a cylinder on a torsion wire immersed in it. The cylinder is deflected and the liquid stirred. Stirring is then stopped and the cylinder released. The torsion in the wire would then vary with time as in Fig. 8 for the two types of material.

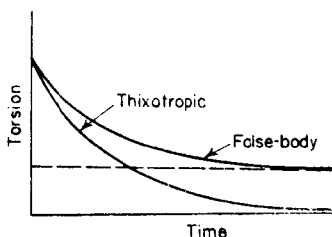


FIG. 8.

With the false-bodied materials there would be a residual torsion in the wire indicating that the material can offer permanent resistance to shear

immediately after stirring, i.e. it retains a finite yield value. A true thixotropic material would show a residual torsion only if some time elapsed after stirring had ceased before the cylinder was released, to allow structure to build-up.

In a like fashion, false-bodied materials can retain elasticity (see Section 1.4). This would result in a recoil of the cylinder in the above experiment.

(b) Rheoplectic fluids—formation of structure by shear

This is a case of gradual formation of structure by shear, whereas so far the properties of structured materials have been explained on the basis that shearing tends to destroy structure.

Freundlich and Juliusberger,⁽⁶⁾ using a 42 per cent gypsum paste ($1 - 10\mu$) in water, found that after shaking, this material re-solidified in 40 min if at rest, but in 20 sec if the vessel was gently rolled in the palms of the hands. This seems to indicate that small shearing motions facilitate structure build-up but large shearing (shaking) destroys it. There is a critical amount of shear beyond which reformation of structure is not induced but breakdown occurs. This behaviour is also observed with dilute aqueous solutions of vanadium pentoxide and bentonite.

There are other materials, however, in which structure only forms under shear and gradually disintegrates when at rest. This is usually termed 'rheopexy' but it is quite distinct from the definition of this term given by Freundlich to the behaviour of gypsum pastes. Even so this behaviour is only found at moderate rates of shear, for if shearing is rapid the structure does not form. A 0.005 N suspension of ammonium oleate behaves in this way. Consider the flow of this material through a capillary tube. At a moderate pressure difference the flow is rapid at first and then decreases as the structure builds up. At a high pressure difference the flow is always rapid and does not fall off because the structure does not build up at high rates of shear.

(c) Relation between time-dependent and time-independent fluids

Thixotropy is rather like pseudoplasticity in which the time required for the alignment of particles is not negligible. This time effect for 'pseudoplastic materials' is not observable in the apparatus normally used for the testing of these fluids. The difference then is only a matter of degree.

In the same way rheoplectic fluids (e.g. ammonium oleate) are superficially similar to their time-independent counterparts (dilatant fluids) in which the time for structure build-up is insignificantly small. Here, however, the analogy is not so close because rheopexy is a case where build-up is brought about by small shearing rates only. There is an upper limit to the shear rate beyond which the analogy breaks down.

1.4 VISCOELASTIC FLUIDS

A viscoelastic material is one which possesses both elastic and viscous properties, i.e. although the material might be viscous, it exhibits a certain elasticity of shape. This concept is perhaps most easily visualized in the case of a very viscous liquid such as pitch. Suppose initially we consider the simplest case where we assume Newton's law for the viscous component and Hooke's law for the elastic component. In steady state flow under a shear stress τ the rate of shear will be τ/μ_0 where μ_0 is a constant Newtonian viscosity coefficient. Suppose now that the shear stress is increased to $\tau + \delta\tau$ very rapidly. The material will now be sheared through an additional angle $\delta\tau/G$ where G is a rigidity modulus. There is therefore an additional rate of shear proportional to the rate of change of stress at any instant and the total rate of shear is given by

$$\dot{\gamma} = \tau/\mu_0 + \dot{\tau}/G \quad [1.4.1]$$

or we can write this as

$$\tau + \lambda_1 \dot{\tau} = \mu_0 \dot{\gamma} \text{ where } \lambda_1 = \mu_0/G \quad [1.4.2]$$

This equation was first proposed by Maxwell,⁽⁶⁾ and liquids which are described by it are usually referred to as 'Maxwell liquids'.

The parameter λ_1 has dimensions of time and it is seen from Eqn. [1.4.2] that it is the time constant of the exponential decay of stress at a constant strain, i.e. if the motion is stopped the stress will relax as $\exp(-t/\lambda_1)$. Consequently λ_1 is known as the relaxation time. Schofield and Scott-Blair⁽⁷⁾ have successfully applied the Maxwell equation to flour doughs.

Oldroyd⁽⁸⁾ investigated the elastic and viscous properties of emulsions and suspensions of one Newtonian liquid in another and derived theoretically the differential equation relating the shear stress τ and the rate of shear $\dot{\gamma}$ in the form

$$\tau + \lambda_1 \dot{\tau} = \mu_0(\dot{\gamma} + \lambda_2 \ddot{\gamma}) \quad [1.4.3]$$

where the constants μ_0 , λ_1 , and λ_2 can be determined in terms of the physical properties of the mixture. In this system the elastic strain energy is stored during flow by virtue of the fact that interfacial tension provides the restoring force which makes the individual drops resist changes of shape. The same equation was also derived by Frölich and Sack⁽⁹⁾ for a dilute suspension of solid particles in a viscous liquid. Elastic strain energy is stored because the solid elastic particles are deformed by the flow of the surrounding liquid.

In this equation the constant μ_0 can be identified as the viscosity at low

rates of shear in the steady state, i.e. when $\dot{\tau} = \dot{\gamma} = 0$. The constant λ_1 is a relaxation time and the physical significance of this is that if the motion is suddenly stopped the shear stress will decay as $\exp(-t/\lambda_1)$; λ_2 is called a 'retardation time' and has the significance that if all stresses are removed the rate of strain decays as $\exp(-t/\lambda_2)$.

Toms and Strawbridge⁽¹⁰⁾ found that the behaviour of dilute solutions of poly-methylmethacrylate in pyridine can be described by means of an equation of this sort. It also describes the behaviour of some bitumens.

It is apparent, then, that a viscoelastic fluid cannot be characterized by a simple rheological equation of the form of $\dot{\gamma} = f(\tau)$. The essential difference is that the rheological equation contains the time derivatives of both τ and γ in general. The general case may be written

$$f_1(D) \tau = f_2(D) \gamma \quad [1.4.4]$$

or alternatively as a polynomial in D

$$\sum_{n=0}^N \alpha_n D^n \tau = \sum_{m=0}^M \beta_m D^m \gamma$$

where D is the differential operator, d/dt .

1.5 MECHANICAL ANALOGIES TO VISCOELASTIC FLUIDS

The rheological equation for a viscoelastic fluid

$$f_1(D) \tau = f_2(D) \gamma$$

is general and if solved subject to the correct boundary conditions will give the response of the material to any imposed stress or strain. However for real fluids the equations are very difficult to solve, even assuming that the values of the relevant parameters can be derived from experiment; but a great deal of qualitative information can be derived from a study of idealized mechanical models or analogies which are designed to duplicate, more or less closely, the observed time-dependence of a real fluid. Their behaviour is more easily visualized than that of a fluid, perhaps especially by engineers. A study of models also suggests a valuable method of characterizing a fluid by a single parameter. This is discussed later in this section.

These models are made up of combinations of springs and dash-pots. The force on a spring is proportional to strain and the force on a dash-pot is proportional to rate of strain. Consequently the springs and dash-pots in a model represent the elastic and viscous properties of the fluid respectively. The basic elements in any mechanical model are a parallel combination of a

spring and dash-pot, known as a Voigt element, and a series combination, known as a Maxwell element (since its equation is the same as that of a Maxwell body discussed previously). These elements represent the behaviour of idealized materials. Real fluids will consist of a more or less complicated combination of these basic elements. The models for complex materials will be derived by first considering the two basic elements and then generalizing them.

(a) Voigt body

The mechanical analogy of a Hookean solid is a spring, and that of a Newtonian liquid is a dash-pot. If we combine these in parallel we have what is known as a Voigt body.

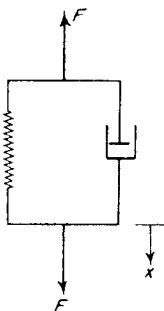


FIG. 9. Voigt body.

The equation of motion of this body is

$$F = k_1 x + k_2 \dot{x}$$

where k_1 is the spring constant and k_2 is the damping constant of the dash-pot.

If we regard the force as analogous to stress and the extension as analogous to strain we could say that this body is a mechanical analogy for a fluid whose behaviour in shear is described by the equation

$$\tau = G\gamma + \mu \dot{\gamma} \quad [1.5.1]$$

where μ is a viscosity and G is a rigidity modulus.

Integrating Eqn. [1.5.1] we get in general

$$\gamma = \exp\left(-\frac{G}{\mu} t\right) \left[\gamma_0 + \frac{1}{\mu} \int \tau \exp\left(\frac{G}{\mu} t\right) dt \right] \quad [1.5.2]$$

where γ_0 is the strain at $t = 0$.

If the stress is constant at τ_0 and the initial strain is zero we have the simplified case

$$\gamma(t) = \frac{\tau_0}{G} \left[1 - \exp(-t/\lambda) \right] \quad [1.5.3]$$

where $\lambda = \mu/G$ is the retardation time. If the stress is removed the strain vanishes exponentially with time constant λ . (This means that the strain falls to $1/e$ of its initial value in time λ .)

It should be noted here that a Voigt body is really a viscoelastic 'solid' since it can be seen that it does not exhibit unlimited non-recoverable viscous flow. It will come to rest in fact when the spring has taken up the load.

(b) Maxwell body

The Maxwell body consists of a spring and dash-pot in series. The equation of this body in rheological terms is

$$\dot{\gamma} = \dot{\tau}/G + \tau/\mu$$

and this is seen to be the same as the Maxwell body considered previously.

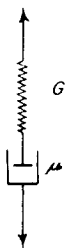


FIG. 10. Maxwell body.

Integrating we get

$$\tau = \exp\left(-\frac{G}{\mu} t\right) \left[\tau_0 + G \int \gamma \exp\left(\frac{G}{\mu} t\right) dt \right] \quad [1.5.4]$$

where τ_0 is the stress at time zero.

If a Maxwell body is subjected at $t = 0$ to a constant strain, γ_0 the stress will decay as

$$\tau(t) = \gamma_0 G \exp(-t/\gamma) \quad [1.5.5]$$

where $\gamma = \mu/G$ is the relaxation time.