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THE STABILITY OF A FILAMENT OF A VISCOELASTIC FLUID

by

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A dissertation submitted to the Graduate Faculty in Engineering in partial fulfillment of the requirement for the degree of Doctor of Philosophy, The City University of New York.

1969

TO NIGI

This manuscript has been read and accepted for the Graduate Faculty in Engineering in satisfaction of the dissertation requirement for the degree of Doctor of Philosophy.

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NOMENCLATURE

The principal symbols are shown below. Symbols which denote quantities which have only a temporary significance are not shown, but are clearly defined in the text. The dyadic notation is briefly discussed in section 1.3. Throughout this work, overhead bars ($\bar{\ }$) are used to denote quantities associated with some primary steady flow, while the superscript (^{o}) is used to denote perturbed quantities. The numbers in parenthesis which occasionally follow the definitions indicate the equation number which corresponds to the particular definition.

$\underset{\sim}{A}^{(N)}$ .... Nth Rivlin-Ericksen kinematic tensor, (1.43)

$\underset{\sim}{B}$ .... Left Cauchy-Green tensor, (1.20)

$\underset{\sim}{C}$ .... Right Cauchy-Green tensor, (1.17)

$\underset{\sim}{C}_t$ .... Right relative Cauchy-Green tensor, (1.46)

C_r .... Residues of the complex viscosity $\hat{\eta}(\alpha)$, (2.3)

$\underset{\sim}{D}$ .... Rate of strain tensor, (1.33)

D/Dt ..... Material time derivative, (1.11)

d_c/dt .... Convected derivative, (1.57)

d^c/dt .... Convected derivative, (1.58)

ds^2 ...... Element of the arc length

det ...... The determinant operator

NOMENCLATURE (continued)

$\underset{\sim}{E}$ .... Eulerian strain tensor, (1.22b)

$\underset{\sim}{E}^{(L)}$....... Linear strain tensor, (1.29)

E .... One-dimensional strain

e .... Extension rate, (5.1)

$\underset{\sim}{F}$ .... Deformation gradient function, (1.13)

$\underset{\sim}{F}_t$ .... Relative deformation gradient function, (1.45)

F_d .... Total external drag, (4.29)

F .... Dimensionless group defined by eq. (4.36)

$\underset{\sim}{G}_t(t-s)$ .. History of the strain, (1.82a)

G .... Dimensionless group defined by eq. (4.36)

$\underset{\sim}{g}$ .... Body force vector

H .... Mean curvature of the filament surface, (3.6)

h .... Elastic modulus, (2.2)

$\underset{\sim}{I}$ .... Identity tensor

$I_n(\ )$ .... Modified Bessel function of the first kind of order n

i .... $\sqrt{-1}$

$\underset{\sim}{J}$ .... Lagrangian strain tensor, (1.23a)

K .... Shear rate

$\underset{\sim}{K}^o(t)$ .... Indefinite integral of $\underset{\sim}{\Gamma}^o(t)$, (5.34)

Kn( ) .... Modified Bessel function of the second kind of order n

NOMENCLATURE (continued)

k .... Wave number of a disturbance-wave

k^* .... Wave number of the fastest growing disturbance-wave

L .... Used throughout to denote some characteristic length

$M(s;e)$ ... Memory Kernels, (5.30)

$\underset{\sim}{n}$ .... Outward unit normal

P,Q ...... Non-linear operators involving time derivatives, (2.21)

P_L, Q_L ... Linear operators in $\partial/\partial t$

p .... Pressure

q_1, q_2 ... Material functions in steady extension, (5.3)

$R(\ )$ ..... Variable radius of the filament

R_o .... Constant radius of the filament (or unperturbed jet)

R_L .... Reynolds No., (4.36)

R_m .... Dimensionless group defined by eq. (4.36)

r .... Radial coordinate

$\underset{\sim}{S}$ .... The stress tensor

$\underset{\sim}{s}$ .... The stress vector

s .... Time

$\underset{\sim}{T}$ .... Extra stress tensor; $\underset{\sim}{T} = \underset{\sim}{S} + p\underset{\sim}{I}$

T_z .... Dimensionless stress defined by eq. (4.77)

T .... Dimensionless stress defined by eq. (4.77)

NOMENCLATURE (continued)

T .... $T_{zz} - T_{rr}$, (5.74a)

$\underset{\sim}{t}$ .... Tangential unit vector, (4.6b)

t .... Time

tr .... The trace operator, (1.3)

$\underset{\sim}{u}$ .... Displacement vector, (1.24)

V_o .... Used to denote a constant or an average velocity

$\underset{\sim}{v}$ .... Velocity vector

v_z .... Axial velocity

v_r .... Radial velocity

v .... Tangential velocity

v .... Dimensionless velocity defined by eq. (4.34)

W .... Mass flow rate, (4.17)

z .... Axial coordinate

Greek Symbols

α........ Complex variable often interpreted as a complex frequency associated with a disturbance-wave

α^* .... The fastest growing rate of a disturbance-wave

β_o, β_1 ... Fluid constants, (4.62)

$\underset{\sim}{\Gamma}$........ Velocity gradient tensor

Δ........ Linear operator defined by eq. (3.14)

δ .... Dimensionless radius defined by eq. (4.34)

NOMENCLATURE (continued)

ζ .... Dimensionless length defined by eq. (4.34)

η_0 .... Zero or Newtonian viscosity

$\hat{\eta}(\alpha)$ ..... Complex viscosity of linear viscoelasticity, (2.3)

$\hat{\eta}(iw)$ .... Complex viscosity in vibrational motions

η', η'' ... The real and imaginary parts of a complex viscosity

$|\hat{\eta}|$ ...... The absolute value of a complex viscosity

η_∞ .... $\lim\limits_{\alpha \to \infty} \hat{\eta}(\alpha)$

$\hat{\eta}(\alpha, K)$ ... Complex viscosity of the superposed motion in steady shear flow

$\hat{\eta}(\alpha, e)$ ... Complex viscosity of the superposed motion in steady extension

η_T .... Trouton, or elongational viscosity, (1.98)

$\varphi(t-s)$ ... Relaxation function of linear viscoelasticity

λ_i .... Fluid time constants

μ_r .... Reciprocals of the relaxation times of linear viscoelasticity, (2.2)

ξ .... Small surface perturbation

ρ .... Density

σ .... Coefficient of surface tension

Σ .... Dimensionless group defined by eq. (4.36); also, in chapter 5 Σ is used to denote the quantity given by eq. (5.96)

NOMENCLATURE (concluded)

τ .... Time

Φ .... Relaxation Kernels, (5.47)

ψ .... Stream function, (3.12)

Ψ_i ...... Relaxation Kernels, (5.51), (5.54)

w .... Angular frequency of a sinusoidal motion.

$\underset{\sim}{\Omega}$ .... Vorticity tensor, (1.34)

Hebrew Symbols

ב .... Dimensionless group defined by eq. (4.70)

ג .... Dimensionless group defined by eq. (4.77)

ל .... Wave length of a disturbance-wave

Other Symbols

$\underset{\sim}{0}$ .... Zero tensor

$\mathfrak{D}/\mathfrak{D}t$ ..... Jaumann derivative, (1.56)

$\partial/\partial t$ ..... Partial derivative

∇ .... The gradient operator

$(\hat{\ })$ ...... Denotes a Laplace transform

$\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ .... Functional, (1.82)

$\delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ ..... Frechet differential, (5.26)

ABSTRACT

The hydrodynamical behavior of a filament of a viscoelastic fluid at rest and under tension constitutes the subject matter of this work.

The stability of a filament of fluid which is initially at rest (relative to small surface disturbances) is studied in reference to the phenomenon of the break-up of capillary jets. The stability analysis shows that a jet of a viscoelastic fluid which is characterized by a complex viscosity that is bounded in absolute value is <u>less</u> stable than a jet of a Newtonian fluid of the same zero viscosity. The theoretical results are subsequently evaluated in the light of existing experimental data.

The steady state behavior of a liquid filament under tension is studied within the context of fiber spinning processes. The results of the theoretical analysis show that fluids which exhibit an apparent viscosity which increases with the axial component of the deformation-rate, are capable of forming filaments (under tension) of lengths greater than the corresponding filaments of fluids which do not possess this property. The, physically observed, superior spinnability of viscoelastic fluids over Newtonian fluids is, hence, attributed to the above

property.

The stability of a cylindrical liquid filament in <u>steady extension</u> (relative to small surface disturbances) is also examined. The rheological properties of the filament fluid are represented by Noll's <u>Simple Fluid Theory</u>. Constitutive equations which describe small perturbations superposed on steady extension are developed. The stability question is, in turn, dependent upon two material functions of the steady motion, and a complex viscosity which arises from the constitutive equations of the perturbed flow. The analysis shows that whereas a filament of a viscoelastic fluid at rest is less stable than a Newtonian filament of the same zero viscosity, the opposite effect is more likely to occur when the filament is under tension (steady extension).

CHAPTER 1

INTRODUCTION

1.1 PRELIMINARY REMARKS

When a Newtonian fluid is ejected from a cylindrical nozzle, the emerging jet is subjected to small surface disturbances. Owing to the action of the surface tension forces these initial disturbances develop into an axisymmetric wave which grows exponentially, the jet finally breaking up into droplets. This break-up phenomenon is illustrated on the following page (fig. 1.1). The break-up pattern of a jet of a typical viscoelastic fluid, on the other hand, is markedly different (fig. 1.2). No wave formation is apparent. Instead, a series of large droplets are formed at irregular intervals along the axis of the jet; these droplets are connected by long cylindrical filaments of fluid which thin with distance, eventually leading to the break-up of the liquid column. These filaments possess a remarkable stability with respect to any theoretical prediction that is based on the behavior of a Newtonain fluid.

The formation of liquid filaments is not typical only to the behavior of capillary jets of viscoelastic fluids. In fact, it seems to represent a unique feature in all atomization processes which involve viscoelastic fluids.

Fig. 1.1 The Breakup of a Newtonian Jet.

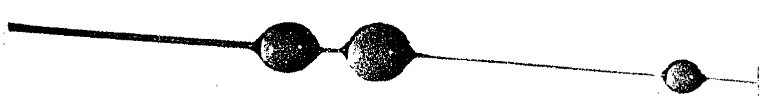

Fig. 1.2 The Breakup of a Viscoelastic Jet.

Dombrowsky [19] has studied the behavior of Napalm solution (a highly elastic fluid) in an expanding sheet formed by a fan jet, and found that instead of atomizing directly into droplets the sheet breaks up into a large number of filaments of fluid. The break-up of a liquid sheet formed by two impinging jets of viscoelastic fluids reveals a similar pattern.

A stabilizing effect of viscoelastic additives on the atomization of fluids has also been noted by Davies [18]. Wilcox [73] has shown that viscoelastic additives increase the stability of liquid droplets exposed to high-velocity air stream and concluded that such additives may find useful applications in various processes in which fluids are atomized. Viscoelastic additives have also been suggested for use in liquid rocket fuel systems to prevent sloshing of the fuel at zero gravity.

The effects noted above are of particular significance since they can be brought about by the addition of only small amounts of viscoelastic additives to a Newtonian fluid without introducing any appreciable changes in other properties of interest (e.g. heat capacity). An understanding of these effects can therefore prove very valuable in many engineering applications such as liquid-liquid extraction, chemical sprays and fuel combustion.

The formation of liquid filaments during the atomization of viscoelastic fluids poses two questions. The first relates to the formation of the filament per se. In contrast to Newtonian fluids, viscoelastic fluids are generally capable of producing elongated liquid filaments. Some of these fluids can be drawn by hand into a stable filament over a yard long and a fraction of a millimeter in diameter. This property represents one of the essential features of the process of fiber spinning. It has been claimed that viscoelastic properties are necessary to the spinning process. Indeed, some of the characteristic viscoelastic phenomena have been adopted as rough measures of spinnability (i.e. the ability to form fibers). However, the apparent adaptability of viscoelastic fluids to the spinning process and its relation to the rheological properties that these fluids possess are not entirely understood. Furthermore, the steady flow behavior of a filament under tension has not yet been translated adequately into a proper mathematical analysis.

The second problem relates to the unusual stability of the filaments once they are formed. Thin threads of fluid which have been occasionally observed during the break-up of a Newtonian jet are very short-lived and soon break into spherical droplets. On the other hand, the corresponding viscoelastic filaments exhibit no apparent instabilities.

This stable behavior may be related to the possibility that the threads are subjected to axial tension in consequence of the relative motion of the large droplets to which the threads are connected. The fact that the threads grow thinner and longer along their flight way seems to confirm such a possibility. Inasmuch as viscoelastic fluids are known to possess a resistance to stretching which increases with the stretching rate, it is likely that a state of axial tension along a filament may enhance its stability. This, however, has never been confirmed and a corresponding quantitative analysis has not yet been undertaken.

1.2 AIM AND SCOPE OF THE THESIS

The hydrodynamical behavior of a filament of a viscoelastic fluid in a relaxed state and under tension constitutes the subject matter of this thesis. A concurrent consideration emerges continuously throghout the work, namely, the constitutive description of the rheological behavior of the filament fluid. This in itself is a major problem since our knowledge of proper constitutive equations for real fluids is rather incomplete. A serious consideration was given to the choice and the subsequent evaluation of the constitutive theories which are used in the present study. This could perhaps be regarded as an additional contribution to the understanding of viscoelastic fluids.

The remainder of this chapter is designed to provide the reader with the necessary background for the subsequent perusal of the main body of work. We review, in this order, (a) the basic concepts and definitions which are used to describe the deformation of a continuum; (b) the basic properties of viscoelastic fluids; (c) constitutive theories which are commonly used to describe the rheological properties of viscoelastic fluids; (d) previous work bearing upon the present study.

Since a considerable portion of the present work involves the use of a linear stability analysis, chapter 2 is devoted to the study of the dynamic response of viscoelastic fluids to small deformations which are superimposed either on a quiescent state or on initially stressed fluids. The linear theory of viscoelasticity can also be used to establish an important guideline for the choice or the formulation of proper non-linear theories.

Chapter 3 present an analysis of the capillary stability of a jet of a viscoelastic fluid. The analysis is based on the assumption that the jet is initially relaxed and is moving horizontally with a constant velocity. It is therefore equivalent to the description of the stability of a cylinderical filament which is initially at rest.

The fourth chapter is devoted to the study of the steady state behavior of a filament under tension. Inasmuch as this represents the mechanical aspect of the spinning of fibers, the problem has been formulated and solved under conditions that are encountered in commercial spinning processes.

Chapter 5 is devoted to the study of the stability of a filament which is subjected to axial tension. Here the basic flow profile which describes the elongation of the filament under tension is assigned a priori, and the stability of the filament, relative to small surface disturbances, is examined by means of the usual perurbation techniques. Chapter 6 closes this work with summary and conclusions.

1.3 KINEMATICS

Kinematics is the description of the motion of a body. Within the domain of continuum mechanics a body is viewed as a set of points (generally referred to as particles or material points) filling a three-dimensional space continuously. The motion of the body is considered as the aggregate of the motion of the individual points. Consequently, the motion of the constituent particles determines the motion of the body as a whole. We review here some of the basic kinematic quantities which are used throughout the present work.

On Notation

Vectors will be denoted by lower case letters with a tilde underneath: $\underset{\sim}{v}$, $\underset{\sim}{u}$,...

Second-order tensors are denoted by upper case letters with a tilde underneath: $\underset{\sim}{S}$, $\underset{\sim}{T}$, ...

For simplicity, vectors and second-order tensors are referred to a cartesian coordinate system unless the specific situation requires otherwise. Whenever necessary the tensorial index notation is used to denote the components of vectors or tensors; i.e. C_{ij}, v_i. The transpose and the inverse of a second-order tensor $\underset{\sim}{A}$ are given by $\underset{\sim}{A}^T$ and $\underset{\sim}{A}^{-1}$ respectively.

The unit tensor, or identity transformation is denoted by $\underset{\sim}{I}$,

$$\underset{\sim}{I}\cdot\underset{\sim}{A} = \underset{\sim}{A} \tag{1.1}$$

$\det\underset{\sim}{A}$ is the determinant operator; i.e.

$$\det\underset{\sim}{A} = |A_{ij}| \tag{1.2}$$

$\mathrm{tr}\underset{\sim}{A}$ shall denote the trace of $\underset{\sim}{A}$,

$$\mathrm{tr}\underset{\sim}{A} = \underset{\sim}{A}:\underset{\sim}{I} \tag{1.3}$$

∇ denotes the gradient operator. Finally, $\underset{\sim}{A}\cdot\underset{\sim}{A}$ is given by $\underset{\sim}{A}^2$.

The deformation of a body can be described by following up the motion of its particles from some reference configuration. Thus, the <u>deformation function</u>

$$\underset{\sim}{x} = \underset{\sim}{x}(\underset{\sim}{\xi}, t) \tag{1.4}$$

relates the position $\underset{\sim}{x}$ of a particle at time t to the position $\underset{\sim}{\xi}$ that the particle has occupied in the reference configuration at time t_o ($t_o < t$). It is assumed that

$$\frac{\partial(x_1, x_2, x_3)}{\partial(\xi_1, \xi_2, \xi_3)} \neq 0 \tag{1.5}$$

so that eq. (1.4) can be inverted to

$$\underset{\sim}{\xi} = \underset{\sim}{\xi}(\underset{\sim}{x}, t) \tag{1.6}$$

A quantity f can be described by a <u>material description</u> involving $\underset{\sim}{\xi}$ and t as the independent variables or, by a <u>spatial</u> description in which $\underset{\sim}{x}$ and t are the independent variables. The two descriptions are interchangeable, that is

$$f(\underset{\sim}{x}, t) = f[\underset{\sim}{\xi}(\underset{\sim}{x}, t), t] \tag{1.7}$$

and

$$f(\underset{\sim}{\xi}, t) = f[\underset{\sim}{x}(\underset{\sim}{\xi}, t), t] \tag{1.8}$$

where the quantities f appearing on both sides of eqs. (1.7) and (1.8) need not be of course identical in their functional form. Associated with these two descriptions are the following time derivatives:

$$\frac{\partial f(\underset{\sim}{x},t)}{\partial t} = \left.\frac{\partial f}{\partial t}\right|_{x\,=\,\text{constant}} \tag{1.9}$$

and the material derivative

$$\frac{Df(\underset{\sim}{\xi},t)}{Dt} = \left.\frac{\partial f}{\partial t}\right|_{\underset{\sim}{\xi}\,=\,\text{constant}} \tag{1.10}$$

The two derivatives are related by means of a chain rule,

$$\frac{Df(\underset{\sim}{x},t)}{Dt} = \frac{\partial f(\underset{\sim}{\xi},t)}{\partial t} = \underset{\sim}{v}\cdot\nabla f + \left(\frac{\partial f}{\partial t}\right)_{\underset{\sim}{x}} \tag{1.11}$$

where $\underset{\sim}{v} = \underset{\sim}{v}(\underset{\sim}{x},t)$ is the velocity given by

$$\underset{\sim}{v} = \frac{D\underset{\sim}{x}}{Dt} \tag{1.12}$$

The gradient of $\underset{\sim}{x} = \underset{\sim}{x}(\underset{\sim}{\xi},t)$ with respect to $\underset{\sim}{\xi}$ is the

<u>deformation gradient</u> $\underset{\sim}{F}$,

$$F_{ij} = \frac{\partial x_i}{\partial \xi_j} \tag{1.13}$$

Eq. (1.5) implies that

$$\det\underset{\sim}{F} \neq 0 \tag{1.14}$$

The square of the element of length at time t is give by

$$ds^2 = d\underset{\sim}{x}\cdot d\underset{\sim}{x} \tag{1.15}$$

or equivalently by

$$ds^2 = d\underset{\sim}{\xi}\cdot(\underset{\sim}{C}\cdot d\underset{\sim}{\xi}) \tag{1.16}$$

where $\underset{\sim}{C}$ is a symmetric, positive-definite tensor often called

the <u>right Cauchy-Green tensor</u> ; it is given by

$$\underset{\sim}{C} = \underset{\sim}{F}^T\cdot\underset{\sim}{F} \tag{1.17}$$

or

$$C_{k\ell} = \frac{\partial x^i}{\partial \xi^k}\frac{\partial x^i}{\partial \xi^\ell} \tag{1.18}$$

Likewise, the squared element of arc at $\underset{\sim}{\xi}$ is given by

$$ds_o^2 = d\underset{\sim}{\xi} \cdot d\underset{\sim}{\xi} = d\underset{\sim}{x} \cdot (\underset{\sim}{B}^{-1} \cdot d\underset{\sim}{x}) \tag{1.19}$$

where $\underset{\sim}{B}^{-1}$ is the inverse of the <u>left Cauchy-Green tensor</u> $\underset{\sim}{B}$:

$$\underset{\sim}{B} = \underset{\sim}{F} \cdot F^T \quad ; \quad \underset{\sim}{B}^{-1} = \underset{\sim}{F}^{T^{-1}} \cdot \underset{\sim}{F}^{-1} \tag{1.20}$$

or

$$B_{k\ell} = \frac{\partial x^k}{\partial \xi^i} \frac{\partial x^\ell}{\partial \xi^i} \quad ; \quad B^{-1}_{k\ell} = \frac{\partial \xi^i}{\partial x^k} \frac{\partial \xi^i}{\partial x^\ell} \tag{1.21}$$

The difference in the squared arc lengths at $\underset{\sim}{x}$ and at $\underset{\sim}{\xi}$ gives a measure of the strain. Accordingly

$$ds^2 - ds_o^2 = 2(d\underset{\sim}{\xi} \cdot \underset{\sim}{J} \cdot d\underset{\sim}{\xi}) = 2(d\underset{\sim}{x} \cdot \underset{\sim}{E} \cdot d\underset{\sim}{x}) \tag{1.22}$$

where

$$2\underset{\sim}{J} = \underset{\sim}{C} - \underset{\sim}{I} \tag{1.23a}$$

and

$$2\underset{\sim}{E} = \underset{\sim}{I} - \underset{\sim}{B}^{-1} \tag{1.23b}$$

are the <u>Lagrangian</u> and the <u>Eulerian</u> strain tensors, respectively.

The classical theory of elasticity usually defines the components of the strain in terms of a displacement vector which describes the positions of a particle at time t_o and at time t. If $\underset{\sim}{\xi}$ and $\underset{\sim}{x}$ are referred to the same coordinate system, the displacement vector $\underset{\sim}{u}$ is given by

$$\underset{\sim}{u} = \underset{\sim}{x} - \underset{\sim}{\xi} \tag{1.24}$$

Now

$$ds_o^2 = d\underset{\sim}{\xi} \cdot d\underset{\sim}{\xi} \tag{1.25}$$

and

$$ds^2 = d\underset{\sim}{x} \cdot d\underset{\sim}{x} = (d\underset{\sim}{u} + d\underset{\sim}{\xi}) \cdot (d\underset{\sim}{u} + d\underset{\sim}{\xi}) \tag{1.26}$$

from which it follows readily that

$$ds^2 - ds_o^2 = \left[\frac{\partial u_k}{\partial x^l} + \frac{\partial u_l}{\partial x^k} - \frac{\partial u^i_k}{\partial x^k}\frac{\partial u^i_l}{\partial x^l}\right] dx^k dx^l \tag{1.27}$$

The Eulerian strain tensor $\underset{\sim}{E}$ can hence be given by

$$E_{kl} = \frac{1}{2}\left[\frac{\partial u_{k}}{\partial x^l} + \frac{\partial u_l}{\partial x^k} - \frac{\partial u^i}{\partial x^k}\frac{\partial u^i}{\partial x^l}\right] \tag{1.28}$$

When the gradient of $\underset{\sim}{u}$, i.e. $\frac{\partial u_k}{\partial x^l}$, is small, corresponding to small deformations, the last terms in eq. (1.28) can be neglected as being of second order of smallness. For small deformation eq. (1.28) reduces to the familiar expression for the linear strain $\underset{\sim}{E}^{(L)}$;

$$E_{kl}^{(L)} = \frac{1}{2}\left(\frac{\partial u_k}{\partial x^l} + \frac{\partial u_l}{\partial x^k}\right) \tag{1.29}$$

Let P and P' be two neighboring particles which at time t are located at points $\underset{\sim}{x}$ and $\underset{\sim}{x} + d\underset{\sim}{x}$, respectively. The velovty at point P relative to P' is given by

$$d\underset{\sim}{v} = \underset{\sim}{\Gamma}^T \cdot d\underset{\sim}{x} \tag{1.30}$$

where $\underset{\sim}{\Gamma}$ is the velocity gradient tensor,

$$\underset{\sim}{\Gamma} = \nabla \underset{\sim}{v} \tag{1.31}$$

$(\nabla \underset{\sim}{v})$ can be decomposed into its symmetric and antisymmetric parts

$$\nabla \underset{\sim}{v} = \frac{1}{2}\left[(\nabla \underset{\sim}{v}) + (\nabla \underset{\sim}{v})^T \right] + \frac{1}{2}\left[(\nabla \underset{\sim}{v}) + (\nabla \underset{\sim}{v})^T \right] \qquad (1.32)$$

The symmetric part is the <u>rate of strain tensor</u> $\underset{\sim}{D}$;

$$\underset{\sim}{D} \equiv \frac{1}{2}\left[(\nabla \underset{\sim}{v}) + (\nabla \underset{\sim}{v})^T \right] \qquad (1.33)$$

the antisymmetric part is the <u>vorticity</u> tensor $\underset{\sim}{\Omega}$;

$$\underset{\sim}{\Omega} \equiv \frac{1}{2}\left[(\nabla \underset{\sim}{v}) - (\nabla \underset{\sim}{v})^T \right] \qquad (1.34)$$

The velocity gradient $\nabla \underset{\sim}{v}$ and the rate of strain tensor are related to the measures of the strain by the following relationships (for proofs see reference 22):

1) $$D\underset{\sim}{F}/Dt = (\nabla \underset{\sim}{v})\cdot \underset{\sim}{F} \qquad (1.35)$$

2) $$D(ds^2)/Dt = 2d\underset{\sim}{x} \cdot (\underset{\sim}{D} \cdot d\underset{\sim}{x}) \qquad (1.36)$$

3) $$D\underset{\sim}{J}/Dt = \tfrac{1}{2} D\underset{\sim}{C}/Dt = \underset{\sim}{F}^T \cdot \underset{\sim}{D} \cdot \underset{\sim}{F} \qquad (1.37)$$

4) $$\left.\frac{D\underset{\sim}{J}}{Dt}\right|_{t=t_o} = \frac{1}{2} \left.\frac{D\underset{\sim}{C}}{Dt}\right|_{t=t_o} = \underset{\sim}{D} \qquad (1.38)$$

since

$$\underset{\sim}{F}(t_o) = \underset{\sim}{F}^T(t_o) = \underset{\sim}{I}$$

5) $$D\underset{\sim}{E}^{(L)}/Dt = \underset{\sim}{D} \qquad (1.39)$$

The higher-order material derivatives of ds^2 define the <u>Rivlin-Ericksen kinematic tensors</u> $\underset{\sim}{A}^{(N)}(\underset{\sim}{x},t)$:

$$\frac{D^N}{Dt^N}(ds^2) = d\underset{\sim}{x}\cdot(\underset{\sim}{A}^{(N)}\cdot d\underset{\sim}{x}) \tag{1.40}$$

from which

$$\underset{\sim}{A}^{(1)} = 2\underset{\sim}{D} \tag{1.41}$$

$$\underset{\sim}{A}^{(2)} = \frac{D\underset{\sim}{A}^{(1)}}{Dt} + (\nabla\underset{\sim}{v})^T\cdot\underset{\sim}{A}^{(1)} + \underset{\sim}{A}^{(1)}\cdot(\nabla\underset{\sim}{v}) \tag{1.42}$$

and

$$\underset{\sim}{A}^{(N)} = \frac{D\underset{\sim}{A}^{(N-1)}}{Dt} + (\nabla\underset{\sim}{v})^T\cdot\underset{\sim}{A}^{(N-1)} + \underset{\sim}{A}^{(N-1)}\cdot(\nabla\underset{\sim}{v}) \tag{1.43}$$

<u>On the Choice of Reference Configuration</u>

In some cases, particularly when the motion of a fluid is described, it is convenient to trace the motion of a particle relative to the position $\underset{\sim}{x}$ which the particle is occupying at the present moment t. The path line of the particle is then given by

$$\underset{\sim}{\xi} = \underset{\sim}{\xi}(\underset{\sim}{x}, \tau) \tag{1.44}$$

where τ is the variable time and $\underset{\sim}{\xi}$ is the position of the particle at time τ. Corresponding to eq. (1.44) one can define, as before, the deformation gradient, the Cauchy-Green and the strain tensors. These are now denoted by $\underset{\sim}{F}_t(\tau)$, $\underset{\sim}{C}_t(\tau)$, etc', and are generally referred to as the <u>relative deformation gradient</u>, the <u>relative right Cauchy-Green tensor</u>, etc'. Thus, for example,

$$F_{(t)ij}(\tau) = \partial\xi^i/\partial x^j \tag{1.45}$$

and

$$\underset{\sim}{C}_t(\tau) = \underset{\sim}{F}_t^{T}(\tau)\cdot\underset{\sim}{F}_t(\tau) \tag{1.46}$$

Further, it is possible to show that $\underset{\sim}{F}(t)$ and $F_t(\tau)$ are related to each other by the expression

$$\underset{\sim}{F}(\tau) = \underset{\sim}{F}_t(\tau)\cdot\underset{\sim}{F}(t) \tag{1.47}$$

1.4 DYNAMICS

The Stress Tensor

Consider a point P in the interior of a body which is subjected to external forces. The resultant contact force acting on a differential area da (with outward unit nornal vector $\underset{\sim}{n}$, fig. 1.3) passing through P is given by $\underset{\sim}{s}\cdot da$, where $\underset{\sim}{s}$ is the stress vector.

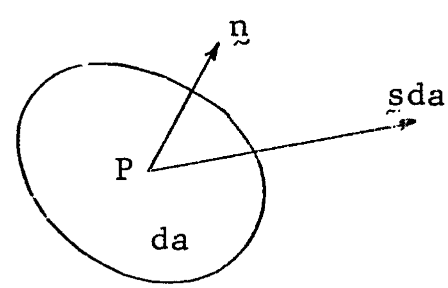

Fig. 1.3 The stress vector

According to the stress hypothesis, the stress vector depends on the orientation of the surface, i.e.

$$\underset{\sim}{s} = \underset{\sim}{s}(\underset{\sim}{x},t,\underset{\sim}{n})$$

However, application of a momentum balance to a small tetrahedron surrounding P shows that there exists a linear transformation, or a tensor $\underset{\sim}{S}$, which transforms the unit vector $\underset{\sim}{n}$ into the stress tensor $\underset{\sim}{s}$,

$$\underset{\sim}{s} = \underset{\sim}{n} \cdot \underset{\sim}{S} \tag{1.48}$$

$\underset{\sim}{S}$ is the stress tensor. Specification of $\underset{\sim}{S}$ as a function of position and time determines the state of stress throughout the body.

By a balance of moments it is, furthermore, possible to show that, in the absence of body moments and couple stresses, the stress tensor is symmetric:

$$\underset{\sim}{S} = \underset{\sim}{S}^T \tag{1.49}$$

For fluids at equilibrium,

$$\underset{\sim}{n} \cdot \underset{\sim}{S} = -p\underset{\sim}{n} \tag{1.50a}$$

or

$$\underset{\sim}{S} = -p\underset{\sim}{I} \tag{1.50b}$$

where p is a hydrostatic scalar pressure. It is customary, therefore, to split the stress tensor into two parts,

$$\underset{\sim}{S} = -p\underset{\sim}{I} + \underset{\sim}{T} \tag{1.51}$$

such that $\underset{\sim}{T} = \underset{\sim}{0}$ ($\underset{\sim}{0}$ being the zero tensor) for fluids at equilibrium. For a compressible fluid, the pressure p is given by the equation of state of the fluid. If the fluid is incompressible, the pressure is indeterminate. This indeterminancy can be removed by adopting some convention such as

$$\mathrm{tr}\underset{\sim}{T} = 0 \tag{1.52}$$

in which case

$$p = -\frac{1}{3}\mathrm{tr}\underset{\sim}{S} \qquad (1.53)$$

and the <u>extra-stress tensor</u> $\underset{\sim}{T}$ coincides with the stress deviator:

$$\underset{\sim}{T} = \underset{\sim}{S} - (\frac{1}{3}\,\mathrm{tr}\underset{\sim}{S})\underset{\sim}{I} \qquad (1.54)$$

<u>The Objectivity of the Stress Tensor</u>

One of the basic postulates of continuum mechanics is the so-called <u>principle of material indifference</u> or, as it is sometimes called, the <u>principle of material objectivity</u> [66] . In general it requires that a response of a material be independent of how it is observed and hence be the same in all frames of reference. In particular it is assumed that the stress tensor, which describes contact forces in a deforming body , be invariant under all changes of frame. If we describe a change of frame $\underset{\sim}{x} \rightarrow \underset{\sim}{x}^*$ by an orthogonal transformation $\underset{\sim}{Q}(t)$ (i.e. $Q^{-1} = Q^{T}$), it follows that the stress tensors corresponding to positions $\underset{\sim}{x}$ and $\underset{\sim}{x}^*$ should satisfy

$$\underset{\sim}{S}^* = \underset{\sim}{Q}\cdot S\cdot \underset{\sim}{Q}^{T} \qquad (1.55)$$

The principle of material objectivity proves to be an important guide in formulating constitutive equations for all materials.

<u>The Stress Flux</u>

In the study of viscoelastic fluids it is often assumed that the rheological behavior of these fluids depends upon the time rate of change of the stress. A tensorial quantity describing the stress rate is commonly called a stress flux. Just as the stress tensor is objective, the stress flux must also be objective. This rules out the ordinary material derivative as a proper stress flux. Instead, the following expressions for the stress flux are commonly used by rheologists:

$$\frac{\mathfrak{D}\underset{\sim}{S}}{\mathfrak{D}t} = \frac{D\underset{\sim}{S}}{Dt} + \underset{\sim}{S}\cdot\underset{\sim}{\Omega} + \underset{\sim}{\Omega}\cdot\underset{\sim}{S} \tag{1.56}$$

$$\frac{d_c\underset{\sim}{S}}{dt} = \frac{D\underset{\sim}{S}}{Dt} + (\nabla\underset{\sim}{v})^T\cdot\underset{\sim}{S} + \underset{\sim}{S}\cdot(\nabla\underset{\sim}{v}) \tag{1.57}$$

$$\frac{d^c\underset{\sim}{S}}{dt} = \frac{D\underset{\sim}{S}}{Dt} - (\nabla\underset{\sim}{v})\cdot\underset{\sim}{S} - \underset{\sim}{S}\cdot(\nabla\underset{\sim}{v})^T \tag{1.58}$$

The operator $\mathfrak{D}/\mathfrak{D}t$ is called the Jaumann derivative or, sometimes, the co-rotational derivative ; it is discussed in detail in reference 44. The operators d_c/dt and d^c/dt are usually called the convected derivatives ; they are discussed in reference 26. It should be pointed out that the definition of the stress flux is not unique. Besides eqs. (1.56)-(1.58) other invariant stress fluxes can be formulated [23].

The Dynamic Equations

Use of the stress tensor leads to the following expression for the balance of linear momentum:

$$\rho\frac{D\underset{\sim}{v}}{Dt} = -\nabla p + \nabla\cdot\underset{\sim}{T} + \rho\underset{\sim}{g} \tag{1.59}$$

where $\underset{\sim}{g}$ is a body force per unit mass. The present work deals almost exclusively with a cylindrical geometry; hence, we give below the components equations of (1.59) referred to a cylindrical coordinate system (r, θ, z) :

$$\rho\left[\frac{\partial v_r}{\partial t} + v_r\frac{\partial v_r}{\partial r} + \frac{v_\theta}{r}\frac{\partial v_r}{\partial \theta} - \frac{v_\theta^2}{r} + v_z\frac{\partial v_r}{\partial z}\right] = -\frac{\partial p}{\partial r}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r\,T_{rr}) + \frac{1}{r}\frac{\partial T_{r\theta}}{\partial \theta} + \frac{\partial T_{rz}}{\partial z} - \frac{T_{\theta\theta}}{r} + \rho g_r \tag{1.60a}$$

$$\rho\left[\frac{\partial v_\theta}{\partial t} + v_r\frac{\partial v_\theta}{\partial r} + \frac{v_\theta}{r}\frac{\partial v_\theta}{\partial \theta} + \frac{v_r v_\theta}{r} + v_z\frac{\partial v_\theta}{\partial z}\right] = -\frac{1}{r}\frac{\partial p}{\partial \theta}$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\,T_{r\theta}) + \frac{1}{r}\frac{\partial T_{\theta\theta}}{\partial \theta} + \frac{\partial T_{\theta z}}{\partial z} + \rho g_\theta \tag{1.60b}$$

$$\rho\left[\frac{\partial v_z}{\partial t} + v_r\frac{\partial v_z}{\partial r} + \frac{v_\theta}{r}\frac{\partial v_z}{\partial \theta} + v_z\frac{\partial v_z}{\partial z}\right] = -\frac{\partial p}{\partial z}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r\,T_{rz}) + \frac{1}{r}\frac{\partial T_{\theta z}}{\partial z} + \frac{\partial T_{zz}}{\partial z} + \rho g_z \tag{1.60c}$$

(In the above, the components of $\underset{\sim}{T}$ and $\underset{\sim}{v}$ are the physical components.)

1.5 SUMMARY OF THE BASIC PROPERTIES OF VISCOELASTIC FLUIDS

The following is a brief summary of the principal phenomena exhibited by viscoelastic fluids. It is designed to acquaint the reader with the type of fluids being considered and to establish some terminology.

A. Shear Thinning Viscosity and Normal Stress Effects in Viscometric Flows

The bulk of the experimental results which characterize the non-linear behavior of viscoelastic fluids has been obtained in a special class of flows known as viscometric flows [15]. These are isochoric steady motions that include

1) Shear flow between parallel plates (simple shear).
2) Tangential flow between two concentric cylinders in relative rotation.
3) Flow in a cylindrical tube.
4) Flow between cone and plate.
5) Helical flow - combined tangential and axial flow in an annulus.

Kinematically, a flow is viscometric if the history of the strain, $\underset{\sim}{C}_t(\tau)$, obeys

$$\underset{\sim}{C}_t(\tau) = \underset{\sim}{I} - (t - \tau)\cdot \underset{\sim}{A}^{(1)}(\underset{\sim}{x},t) + \frac{(t - \tau)^2}{2} \underset{\sim}{A}^{(2)}(\underset{\sim}{x},t) \qquad (1.61)$$

where $\underset{\sim}{A}^{(1)}$ and $\underset{\sim}{A}^{(2)}$ are the first and second Rivlin-Ericksen tensors.

Consider, for example, the steady flow of viscoelastic fluids between two parallel plates. The velocity profile is given by

$$V_1 = K x^2 \ ; \quad V_2 = V_3 = 0 \qquad (1.62)$$

where x^1 is orthogonal to the flow direction x^2, and K is the shear rate. Viscoelastic fluids generally exhibit the following stress pattern,

$$\begin{aligned} S_{12} &= K\,\eta(K) \\ S_{22} - S_{33} &= \sigma_1(K^2) \\ S_{11} - S_{33} &= \sigma_2(K^2) \\ S_{23} = S_{13} &= 0 \end{aligned} \qquad (1.63)$$

where $\eta(K)$ is the shear viscosity, and $\sigma_1(K^2)$ and $\sigma_2(K^2)$ are the first and second normal stress differences, respectively. An equivalent stress pattern is shown in all the viscometric flows. In a Newtonian fluid there are no normal stress differences and, in addition, the viscosity is independent of the shear rate. The viscosity of viscoelastic fluids, on the other hand, is shear dependent and generally decreases with the shear rate; hence the use of the term <u>shear thinning</u> to describe this phenomenon.

The normal stresses in viscoelastic fluids have been studied experimentally in great detail [15]; the results showing that

$$\sigma_2 >> \sigma_1 > 0 \qquad (1.64)$$

The occurence of normal stress differences in the vicinity of free boundaries, such as the edges of cylinders and pipes, causes two striking phenomena which are totally absent in the

the case of Newtonian fluids:

Weissenberg Climbing Effect [72].

When a cylindrical rod is placed in the center of a rotating cylinder, the fluid tends to climb up the rod. This is shown schmatically in fig. 1.4.

Swelling of a Jet.

When a viscoelastic fluid is ejected from a cylindrical nozzle, the diameter of the resulting jet increases considerably before reaching a constant value. This is illustrated in fig. 1.5.

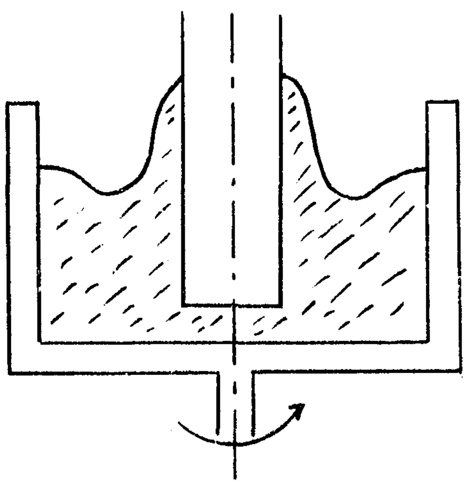

Fig. 1.4 The climbing effect.

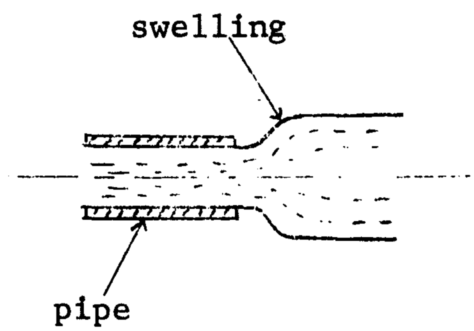

Fig. 1.5 Jet swelling.

B. Stress Relaxation

When a deformed body is brought to rest and held there , the stresses attain their equilibrium value. In a Newtonian fluid the stresses generated during the deformation relax instantaneously, the equilibrium stress being a hydrostatic pressure. A viscoelastic fluid, on the other hand, is characterized by a gradual decay (occuring over a finite amount of time) of the stresses when the fluid is suddenly brought to rest.

C. Recoil

Recoil is the partial recovery of the strain occuring in a deformed viscoelastic fluid when the driving force which causes the motion is suddenly removed. For example, tracers injected into a viscoelastic fluid flowing in a cylindrical tube recoil back when the applied pressure gradient is suddenly released. The jet swelling phenomenon might also be, in part, a manifestation of strain recovery.

D. Drag Reduction

The addition of viscoelastic additives to a Newtonian fluid may result in a considerable reduction of the drag in turbulent flow [30, 57]. The pressure required to produce a given flow rate has often been reduced by as much as 60%. This phenomenon is all the more remarkable, because the additives prove effective even at very low concentrations where the viscosity of the solution hardly differs from that of the solvent.

E. Elongational Viscosity

When a filament of a viscoelastic fluid is elongated due to the application of tensile forces at the filament ends, the apparent viscosity is found to increase with the elongation rate. This is in contrast to the behavior of viscoelatic fluids in viscometric flows in which the shear viscosity decreases with the shear rate. The behavior of a filament of a viscoelastic fluid under tension is considered in detail in chapters 4 and 5.

F. Oscillatory Response of Viscoelastic fluids.

When a Newtonian fluid is subjected to a sinusoidal shearing motion of small amplitude, the resulting, periodic, stress is in phase with the rate of oscillation. In the case of viscoelastic fluids, the stress lags behind the oscillation rate; the phase angle, ϕ , being in the range of $0^\circ < \phi < 90^\circ$, with $\phi = 90^\circ$ corresponding to the response of a purely elastic body. The response of viscoelastic materials to small oscillatory deformation is treated in chapter 2.

Finally we point out that there exists a class of non-Newtonian fluids which exhibit a shear dependent viscosity in viscometric flows but fail to exhibit any of the time-dependent phenomena discussed above (e.g. stress relaxation and recoil). These fluids are commonly called visco<u>in</u>elastic.

1.6 CONSTITUTIVE EQUATIONS

The constitutive equation relates the stresses in a body to its motion. Together with the principles of conservation of mass, momentum and energy it forms a determinate system which, in principle, should be sufficient to furnish a complete solution subject to physically reasonable boundary and initial conditions. The constitutive equation does not represent a physical law; rather, it is a mathematical statement which embodies out total experience with a given material whose rheological properties we wish to describe. A sufficiently general constitutive equation should therefore be capable of reproducing accurately physical phenomena that have been observed in the laboratory. However, once it is written down, the constitutive equation may be regarded as an independent mathematical theory which need not be confined to the limited range of experimental phenomena which guided its formulation. The constitutive equation thus possesses a range which contains but also extends beyond our physical experience. Indeed, since our understanding of the rheological behavior of real substances is often vague and incomplete, the correspondence of actual physical events to those predicted by the constitutive equation is not always clear. In this sense, a constitutive equation defines an ideal material and is very much analogous to the equation of state of classical thermodynamics.

This section is not intended to provide an exaustive review of constitutive equations, nor is it unduly condensed. The engineer as well as the applied theoretician who deal with processes involving viscoelastic fluids are constantly faced with the fundamental problem of selecting a proper constitutive equation which is capable of describing the properties of the medium. It is not surprising therefore, that the literature of the last decade contains virtually hundreds of publications which deal directly or indirectly with constitutive equations. Notwithstanding, the number of hydrodynamical problems for which an exact, or even an approximate solution exist is rather small; and these problems generally belong to the realm of simple geometries and flow situations. Inasmuch as the present work deals with flow situations which have received little attention elsewhere, we review here some of the fundamental concepts which are common to all constitutive theories, and survey the more common constitutive equations that are used by rheologists today.

For a constitutive equation to represent a material adequately, certain physical and mathematical principles must be satisfied. A full discussion of these principles is given by Truesdell [68] ; we summarize some of these below:

(1) Consistency ; the constitutive equation must be compatible with the general principles of balance of mass, momentum

and energy, and the second law of thermodynamics [13].

(2) <u>Coordinate invariance</u> ; the constitutive equation must be stated in a manner that is independent of the specific choice of coordinate system, i.e. in a tensorial language.

(3) <u>The principle of determinism</u> ; the present stress at a material point is determined by the history of the motion in the immediate neighborhood of this point.

(4) <u>The principle of objectivity</u> ; since the response of any material is independent of the observer, the constitutive equation must be invariant with respect to any arbitrary rigid rotation of the frame of reference. That is to say, if the reference frame and the material are rotated at the same time, the constitutive equation remains the same. This principle has already been mentioned in sec. 1.4.

To these principles, we feel, two more guidelines should be added:

(a) The constitutive equation which describes the response of any isotropic viscoelatic material to small perturbations in the vicinity of equilibrium has been deduced by Biot [5] from thermodynamical considerations. Since the validity of this equation is universally acknowledged, it is reasonable to require that any non-linear theory be compatible with the infinitesimal theory of viscoelasticity. That is, when linearized about a quiescent state, any non-linear constitutive equation should reduce to an equation whose time-dependent behavior is equivalent to that predicted by the infinitesimal theory. The consequences of this additional criterion are discussed in detail in chapter 2.

(b) The second guideline is perhaps pertinent to engineering rheology only. It is dictated by the need for explicit constitutive equations that are simple enough to allow a mathematical solution of design problems. This is not to say that the behavior of real materials is necessarily governed by simple constitutive equations. This is to suggest that the behavior of real materials be approximated, if necessary, by equations which allow meaningful correlation of experimental data, and determine dimensionless groups for scale-up purposes.

<u>On the Concept of Fluidity</u>

Much of this work deals with incompressible fluids. Since the intuitive notion of fluidity is often misleading, we adopt here Noll's definition of an <u>incompressible simple fluid</u> [42]; an incompressible simple fluid is a substance which satisfies the following two postulates:

1) The present stress in an incompressible simple fluid is determined, to within a hydrostatic pressure, by the history of the deformation.

2) No preferred configuration exists in a simple fluid. The mathematical formulation of these concepts will be

considered presently. In the meantime we point out that all the fluids considered in the present work, including Newtonian, are special cases of simple fluids. Furthermore, it can be shown that the definition of an incompressible simple fluid encompasses some of the previous notions of fluidity, e.g.
1) A fluid is a substance capable of flowing in all directions.
2) At equilibrium, the stress in a fluid reduces to a hydrostatic pressure.

The first mathematical description of viscoelastic phenomema were based on the superposition of elastic and viscous effects. Maxwell, in his classical paper on the kinetic theory of gases [34], describes the behavior of a linear one-dimentional fluid by

$$S + \lambda \frac{dS}{dt} = 2\eta_0 \frac{dE}{dt} \tag{1.65}$$

where S = S(t) and E = E(t) denote the one-dimensional stress and strain respectively. The mechanical response of the so-called <u>Maxwell fluid</u> can be represented by a spring (elastic) and a dash-pot (viscous) elements in series,

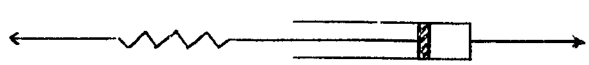

Fig. 1.6 Maxwell element

The first linear one dimensional theory describing a viscoelastic solid was developed by Meyer [38] and was studied in detail by Voigt [70] whose name it usually bears. The

Voigt solid is described by

$$S = hE + \eta_\infty \frac{dE}{dt} \qquad (1.66)$$

and can be represented by a combination of a spring and a dash-pot in parallel,

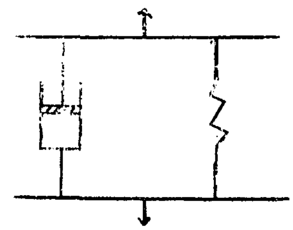

Fig. 1.7 The Voigt element

The Maxwell model can describe the phenomenon of stress relaxation and recoil (λ being the relaxation time, i.e. the time required for the stress to decay to 1/e of its original value). The Voigt solid can exhibit an effect associated with viscoelastic solids, namely, creep [24].

Since the time of the original works of Maxwell and Meyer (corresponding, respectively, to the years 1867 and 1874), a vast literature has developed in which the behavior of viscoelastic materials is represented by mechanical spring and dash-pot elements in all conceivable combinations. Needless to say, such theories apply only to the response of materials to linear (small) deformations. The transition from one-dimensional to the corresponding three-dimensional description has been, on the whole, accomplished arbitrarily by replacing the uni-axial stress and strain by their tensorial counterparts, and substituting partial derivatives for the ordinary derivatives. An incompressible Maxwell fluid thus becomes, for example,

$$\underset{\sim}{T} + \lambda \frac{\partial \underset{\sim}{T}}{\partial t} = 2\eta_0 \underset{\sim}{D} \qquad (1.67)$$

where $\underset{\sim}{T}$ is the deviatoric stress and $\underset{\sim}{D}$ is the rate of strain tensor.

To allow more freedom in correlating linear experimental data, two general model representations have been commonly used. The general Maxwell model (a large number of Maxwell elements, a spring and a dash-pot all connected in parallel, fig. 1.8), and the generalized Voigt model (a large number of Voigt elements, a spring and a dash-pot in series, fig. 1.9).

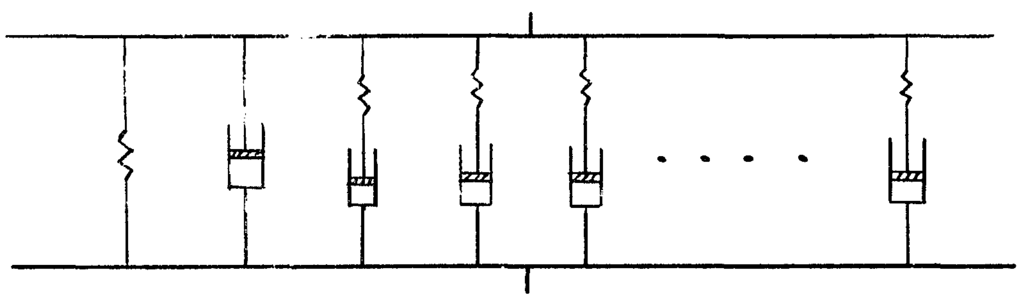

Fig. 1.8 Generalized Maxwell model.

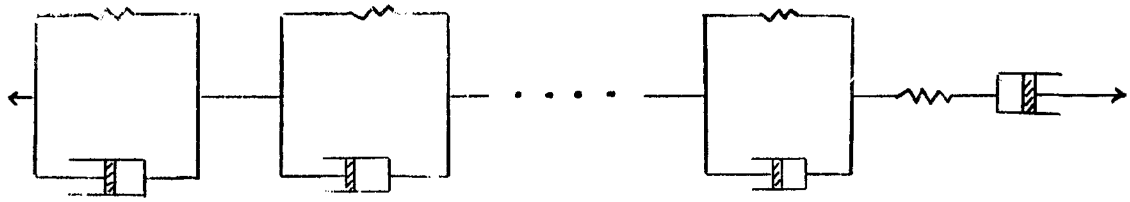

Fig. 1.9 Generalized Voigt model.

The mathematical description of the generalized Maxwell and Voigt materials is generally given in an operator form

$$P_L(\underset{\sim}{T}) = Q_L(\underset{\sim}{E}^{(L)}) \tag{1.68}$$

where P_L and Q_L are polynomials with constant coefficients in $\partial/\partial t$, and $\underset{\sim}{E}^{(L)}$ is the linear strain tensor.

Alternatively, the linear response of viscoelastic materials has been represented by the linear accumulative theory of Boltzmann [7] in which the stress is related to the strain rate by an hereditary integral of the form,

$$\underset{\sim}{T} = 2\int_{-\infty}^{t} \varphi(t-\tau)\underset{\sim}{D}(\tau)d\tau \tag{1.69}$$

For concreteness, the relaxation function $\varphi(t)$ has been generally derived from the same mechanical models discussed above. Thus for a Maxwell fluid

$$\varphi(t) = \frac{\eta_0}{\lambda} e^{-t/\lambda} \tag{1.70}$$

For an incompressible Newtonian fluid

$$\varphi(t) = \delta(t) \tag{1.71}$$

where $\delta(t)$ is the Dirac delta function.

Although the results obtained from the mechanical model representation turn out to be satisfactory, they are not based on any firm theoretical ground. The relevancy of the superposition of springs and dash-pots has been a priori assumed, and the subsequent generalization to three-dimensional materials has been carried out arbitrarily. The same results, however, has been deduced by Biot [5] from thermodynamical considerations which are independent of any postulates as to the nature of the materials or their internal structure. A brief outline of Biot's theory is presented at the beginning of chapter 2 which deals with the response of viscoelastic materials to small deformations.

The first constitutive theories capable of predicting non-linear effects in viscometric flows were formulated as logical extensions to the linear theory of viscoelasticity. Zaremba [74], Oldroyd [43], Metzner [36] and others developed equations which, in analogy to the linear theory, can be represented in an operator form

$$P(\underset{\sim}{T}) = Q(\underset{\sim}{D}) \tag{1.72}$$

where P and Q are polynomials in non-linear time derivative operators such as $\mathfrak{D}/\mathfrak{D}t$, d_c/dt or d^c/dt (see eqs. 1.56 - 1.58). The resulting equations are commonly classified as <u>theories of the rate type</u>. The incompressible Maxwell fluid has thus been generalized to any of the following:

$$\underset{\sim}{T} + \lambda \frac{\mathfrak{D}\underset{\sim}{T}}{\mathfrak{D}t} = 2\eta_0 \underset{\sim}{D} \tag{1.73}$$

$$\underset{\sim}{T} + \lambda \frac{d_c \underset{\sim}{T}}{dt} = 2\eta_0 \underset{\sim}{D} \tag{1.74}$$

$$\underset{\sim}{T} + \lambda \frac{d^c \underset{\sim}{T}}{dt} = 2\eta_0 \underset{\sim}{D} \tag{1.75}$$

As was pointed out before, the choice of the stress flux is not unique. Besides those appearing in eqs. (1.73) - (1.75), other possible invariant time fluxes can be used. However, inasmuch as the properties of materials are independent of the choice of flux, constitutive equations of the rate type should be regarded as descriptions of different fluids [67]. Indeed, each of eqs. (1.73)-(1.75) predicts a different stress pattern in viscometric flows and none is entirely adequate. To achieve a better description of the stress distribution in viscometric flows, other terms has been added to these equations, often, in an arbitrary manner, e.g. Oldroyd 8-constant fluid [43],

$$(1 + \lambda_1 \eth_{abc}) \tilde{T} = 2\eta_0 (1 + \lambda_2 \eth_{a'b'c'}) \tilde{D} \qquad (1.76)$$

where the operator $\eth_{abc}$ is given by

$$\eth_{abc}(\tilde{A}) = \frac{\mathfrak{D}\tilde{A}}{\mathfrak{D}t} - a(\tilde{D}\cdot\tilde{A} + \tilde{A}\cdot\tilde{D}) + b(\tilde{D}:\tilde{A})\tilde{I} + c(\mathrm{tr}\tilde{A})\tilde{D} \qquad (1.77)$$

and where λ_1, λ_2, a, b, c, a', b', c' are material constants. Eq. (1.76) can account for the two normal stress differences and the shear dependent viscosity in viscometric flows.

In 1945, Reiner [52] proposed a different constitutive theory in which the stress, in an incompressible fluid, depends on the rate of strain but, in contrast to the Newtonian theory, the dependence need not be linear,

$$\tilde{T} = \tilde{F}(\tilde{D}) \qquad (1.78)$$

where $\underset{\sim}{F}$ is an isotropic tensor-valued function of $\underset{\sim}{D}$ †. A representation theorem for isotropic functions of symmetric tensors [54], shows that eq. (1.78) can be written as

$$\underset{\sim}{T} = 2\beta_0 \underset{\sim}{D} + 4\beta_1 \underset{\sim}{D}^2 \tag{1.79}$$

where β_0 and β_1 are functions of II and III, two of the principal invariants of $\underset{\sim}{D}$, defined by

$$\begin{aligned} II &= \operatorname{tr} \underset{\sim}{D}^2 \\ III &= \det \underset{\sim}{D} \end{aligned} \tag{1.80}$$

Because of the later work of Rivlin [53], eq. (1.79) has come to be known as the Reiner-Rivlin model. Since the stress in eq. (1.79) depends only on the instantaneous value of the rate of strain, a fluid obeying this equation can not exhibit the time-dependent behavior characteristics of viscoelastic fluids. For this reason, the Reiner-Rivlin fluid is classified as viscoinelastic. Furthermore, while eq. (1.79) can account for non-linear viscosity in viscometric flows, it does not describe correctly the normal stress differences exhibited by real viscoelastic fluids.

A different approach to the constitutive theory of viscoelastic fluids was taken by Rivlin and Ericksen [54], who considered the stress in an incompressible fluid to be a function

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† The components of an isotropic tensor-valued function $\underset{\sim}{F}$ are invariants under rigid rotation of the coordinate system.

of the instantaneous values of the rate of strain and its higher-order time derivatives. Invariance requirements give rise to the following expression

$$\underset{\sim}{T} = \underset{\sim}{F}\left[\underset{\sim}{A}^{(1)}, \underset{\sim}{A}^{(2)}, \underset{\sim}{A}^{(3)} \ldots \underset{\sim}{A}^{(N)}\right] \tag{1.81}$$

where $\underset{\sim}{A}^{(N)}$ is the Nth Rivlin-Ericksen kinematic tensor. Constitutive equations of this type are commonly classified as <u>theories of the differential type</u>. The Rivlin-Ericksen fluid can exhibit all the non-linear effects observed in viscometric flows. On the other hand, the time-dependent behavior of a Rivlin-Ericksen fluid (with $N < \infty$) is not compatible with the infinitesimal theory of viscoelasticity. Indeed, in chapter 2 we shall show that constitutive equations of the differential type involving only a finite number of derivatives of the strain rate should not, in general, be regarded as proper constitutive theories.

All the fluid constitutive equations discussed above are special cases of a more general constitutive theory which is capable of describing the stress pattern in viscoelastic fluids for all motions investigated experimentally. This is <u>Noll's theory of a simple fluid</u> [42]. The essence of the theory is contained in the definition of an incompressible simple fluid given at the beginning of this section. Mathematically,

$$\underset{\sim}{T}(\underset{\sim}{x}, t) = \mathop{\mathfrak{F}}_{s=0}^{s=\infty}\left[G_t(t-s)\right] \tag{1.82}$$

where

$$\underset{\sim}{G}_t(t-s) = \underset{\sim}{C}_t(t-s) - \underset{\sim}{I} \qquad (1.82a)$$

is the relative history of the strain, and is a measure of the strain of the configuration at time t-s ($s \geqslant 0$) relative to the configuration at time t. $\underset{\sim}{\mathfrak{F}}_{s=0}^{s=\infty}$ is a functional - an operator which maps tensor-valued functions unto tensors. Time-dependent phenomena such as stress relaxation have been incorporated into the theory by application of the principle of fading memory [11]. Briefly, this principle asserts that deformations which occured in the distant past should have less effect on the present value of the stress than deformations which occured in the recent past.

In slow motions the behavior of an incompressible simple fluid can be represented by a series of approximations. These approximations are carried out by "retarding" the history of the strain by a factor ϵ such that $0 < \epsilon < 1$. This is equivalent to viewing the history as being the same but occuring at a slower rate. Eq. (1.82) is then expressed as a power series in ϵ. The resulting equation, corresponding to a specific power ϵ^n, is said to be the constitutive equation of the nth-order fluid. The zeroth-order approximation corresponds to a perfect fluid, while the Newtonian fluid represents the first-order approximation to an incompressible simple fluid in slow flows. The next approximation, the so-called second-order fluid, has the following constitutive equation

$$\underset{\sim}{T} = \eta_0 \underset{\sim}{A}^{(1)} + \eta_1 \underset{\sim}{A}^{(1)} \cdot \underset{\sim}{A}^{(1)} + \eta_2 \underset{\sim}{A}^{(2)} \qquad (1.83)$$

where η_0, η_1 and η_2 are material constants. Higher-order approximations have also been considered [55].

The constitutive equation of the second-order fluid has been for many years, including the present, the favorite of many rheologists. The interest in the second-order fluid lies in its simplicity. Together with the momentum equations it can provide solutions to several problems which otherwise are mathematically intractable for the more general constitutive equations. In viscometric flows, the second-order fluid is capable of exhibiting normal stress differences while the viscosity remains constant at a value of η_0. However, unsteady flow solutions involving the second-order fluid generally lead to physically unrealistic results. A considerable literature has been devoted to the study of the stability and the uniqueness of solutions obtained via the constitutive equation of the second-order fluid (e.g. refs. 16, 31, 63). The results of these studies rule out the validity of the second-order theory for any flows other than those which are sufficiently slow. This is not surprising; the constitutive equation of the second-order fluid is not compatible with the infinitesimal theory of viscoelasticity; hence, granting its suitability as an approximate theory for slow flows, on its own right the second-order theory can not be regarded as a proper constitutive theory. This point is taken up in more detail in chapter 2.

For <u>small deformations</u>, which are not necessarily slow, the simple fluid theory can be approximated by a series of integral relations involving the relative history of the strain and memory functions [11]. The first two terms are given by

$$\underset{\sim}{T} = \int_0^\infty m(s)\underset{\sim}{G}_t(t-s)ds + $$
$$+\iint_{s,r=0}^{\infty}\left[a(s,r)\underset{\sim}{G}_t(t-s)\cdot\underset{\sim}{G}_t(t-r)+b(s,r)\underset{\sim}{G}_t(t-s)tr\underset{\sim}{G}_t(t-r)\right]dsdr \tag{1.84}$$

where m(s), a(s,r) and b(s,r) are memory functions. For infinitesimal deformations, eq. (1.84) reduces to Boltzmann equation of infinitesimal viscoelasticity:

$$\underset{\sim}{T} = \int_0^\infty 2\varphi(s)\,\underset{\sim}{D}(t-s)\,ds \tag{1.85}$$

where the relaxation function $\varphi(s)$ is related to m(s) appearing in eq. (1.84) by

$$2\frac{d\varphi(s)}{ds} = -m(s) \tag{1.86}$$

Several constitutive equations similar to eq. (1.84) have been developed by Pao [46], Bougue [9], Bernstein, et al., [4], Tanner [60] and others. These are commonly classified as <u>theories of the integral type</u>.

In addition to the above constitutive equations, several other theories have been proposed in the literature. Such a diversity of approach is created by the constant search for theories that are compatible with our present experience; theories which can pave the way to unexplored areas of investigation. As new physical and theoretical evidence unfolds, some of the existing theories are exposed in their inadequacy, and a new approach has to be selected or deduced. The new theories are not simple, and even the simplest flow situation often constitutes a mathematically formidable task. In fact, modern rheology is characterized by the "backward approach"; that is, one assigns a velocity profile a priori and then proceed to determine whether or not such a flow is dynamically possible. This, as opposed to the "direct approach" in which the system of dynamic equations (constitutive and balance equations) is solved for the velocity distribution under suitable initial and boundary conditions. In view of the complexity of the constitutive equations it is indeed doubtful whether the direct approach can in general be feasible. Finally, regarding the great number of constitutive equations from which the engineer is to select a particular approach; their relative merits must ultimately be judged in relation to the physical evidence and the specific problem of interest.

1.7 REVIEW OF PREVIOUS WORK

The stability of a cylindrical jet of a Newtonian fluid has been studied extensively both theoretically and experimentally [28, 40, 50, 51, 64, 71]. The corresponding problem for a viscoelastic fluid is relatively unexplored. Most of the analytical solutions for a Newtonian jet stem from the pioneering works of Rayleigh and Weber. Rayleigh [50], considered the capillary instability of a cylindrical filament of an inviscid fluid initially at rest and has shown that the rate of growth of the most rapidly growing disturbance-wave is given by

$$\alpha^*_{in} = \frac{1}{2}\sqrt{\frac{\sigma}{2\rho R_o^3}} \tag{1.87}$$

where σ is the coefficient of surface tension and ρ the density of the fluid; R_o is the unperturbed radius of the filament.

In 1931, Weber [71] obtained the complete linear solution for a cylidrical jet of a Newtonian fluid of viscosity η_o. In the absence of interactions with the surrounding atmosphere and with the jet assumed to be initially relaxed, Weber found that the fastest growing disturbance grows at the rate of

$$\alpha^*_o = \frac{\sqrt{\frac{\sigma}{2\rho R_o^3}}}{2 + \frac{3\eta_o}{\rho}\sqrt{\frac{2\rho}{\sigma R_o}}} \tag{1.88}$$

The wave number corresponding to this disturbance-wave is

$$k_o^* = \left[3\eta_o R_o \sqrt{\frac{2R_o}{\rho\sigma}} + 2R_o^2 \right]^{-1/2} \qquad (1.89)$$

An experimental measure of the stability of the jet is the <u>break-up length</u>, L_o, defined as the length of the coherent portion of the jet; it is given by

$$L_o = \frac{C V_o}{\alpha_o^*} \qquad (1.90)$$

where C is a proportionality constant which depends on the initial magnitude of the disturbances, and must be determined experimentally. V_o is the uniform velocity of the jet. From eq. (1.88) one obtains

$$L_o = 2R_o C \left[We^{1/2} + 3\,We/Re \right] \qquad (1.91)$$

where $We = 2 \cdot R_o \cdot V_o^2 \rho/\sigma$ and $Re = 2 \cdot R_o \cdot V_o \rho/\eta_o$ are the Weber and the Reynolds numbers of the unperturbed jet.

Experimental results for laminar Newtonian jets match Weber's theoretical predictions remarkably well [28]. The value of the constant C in eq. (1.91) has been reported in the range of 12.5-19.

An analysis of the stability of a laminar jet of a viscoelastic fluid has been published in 1965 by Middleman [39]. Middleman considered a fluid obeying the following constitutive equation:

$$\underset{\sim}{T} + \lambda_1 \frac{\partial \underset{\sim}{T}}{\partial t} = 2\eta_0 \left(\underset{\sim}{D} + \lambda_2 \frac{\partial \underset{\sim}{D}}{\partial t} \right) \qquad (1.92)$$

where λ_1 and λ_2 are fluid time constants. By a linear analysis which closely parallels Weber's corresponding solution for a Newtonian jet, Middleman obtained an approximate solution for the case in which both λ_1 and λ_2 are very small. His final result is expressed by a ratio of the break-up length L of a viscoelastic jet to the break-up length L_0 of a Newtonian jet of viscosity η_0,

$$\frac{L}{L_0} = 1 - \frac{(\lambda_1 - \lambda_2)}{R_0} \frac{V_0}{(We^{1/2} + 3We/Re)} \qquad (1.93)$$

Since for real fluids $\lambda_1 > \lambda_2$, Middleman concluded that a jet of a viscoelastic fluid obeying eq. (1.92) is less stable than a jet of a Newtonian fluid of viscosity η_0.

Experimental results [27] show that there is a marked difference in the break-up and the shape of viscoelastic jets as compared to Newtonian jets. The basic differences have been illustrated by figs. 1.1 and 1.2. In the case of a jet of a Newtonian fluid, once an axisymmetric wave appears on the surface of the jet, it grows exponentially leading to the formation of droplets whose average diameter matches the wave length corresponding to that predicted by eq. (1.89). In the case of a jet of a typical viscoelastic fluid, no sinusoidal wave formation is apparent. Instead, instability is characterized by the rapid formation of large droplets of fluid which appear irregularly spaced along the axis of the jet. The droplets are connected by long filaments of fluid that thin continuously with the distance and possess a remarkable stability which can not be accounted for by Weber's theory for a Newtonian fluid or by Middleman's corresponding solution for a fluid obeying eq. (1.92). Furthermore, unlike the case of a Newtonian jet which exhibits a break-up length that is unambiguous, reproducible and readily measurable from photographs, the break-up of a viscoelastic jet appears to follow no regular pattern and actual break-up of the liquid column generally occurs due to disruption of the thin filaments.

The theoretical results of Middleman fail to describe the break-up pattern of viscoelastic jets. In particular, they fail to account for the formation and the unusual stability of the long thin filaments observed experimentally. It is, therefore, of interest to investigate if Middleman's results are only limited to the specific viscoelastic model chosen (eq. 1.92) or whether they are, indeed, representative of a larger, more general, class of constitutive theories describing viscoelastic fluids. The present treatment (chapter 3) extends the analysis and encompasses the response of any viscoelastic material to infinitesimal disturbances.

The formation and the unusual stability of the thin filaments of fluid formed during the break-up of a viscoelastic jet, might represent a non-linear phenomena which lie outside the range of the linear analysis of the Weber's type. In fact, high-speed motion pictures [27] indicate that the thin threads connecting the large droplets are subjected to axial deformation caused by the relative motion of the droplets. The filaments are, therefore, not relaxed and their stability relative to small disturbances might strongly depend on the primary flow pattern in the filaments. An analysis of the stability of a filament of fluid, under axial tension, relative to small surface disturbances, might hence provide a considerable insight into this problem. To my knowledge, such an analysis has never been undertaken. Indeed, a complete rigorous solution to this problem might prove to be a formidable task. To begin with, the primary flow behavior of the filament under tension must first be determined or approximated in a suitable way. Further, the response of viscoelastic fluids to small perturbations superimposed upon finite deformations is a subject which is relatively unknown, and only recently has received some due attention. In chapter 2 we examine some of the conclusions which can be drawn from the existing work on the dynamic behavior of viscoelastic fluids undergoing steady flow relative to small superposed perturbations. In chapter 5, a simplified analysis of the stability of a filament of fluid under tension (with respect to small surface disturbances) is presented.

Turning now to the formation of the filaments per se. Three general problems, relevant to this phenomenon, have been considered in the literature: The falling of a fluid under its own wieght, the steady extension of a filament of fluid and the spinning of fibers. Common to all these problems is the consideration of the shape and the flow behavior of a filament of fluid subjected to tensile stress.

In 1906, Trouton [65] published a theoretical and experimental study on the behavior of a fluid falling vertically under its own wight through a round hole. In particular, Trouton sought to measure the elongational viscosity (defined as the ratio of the tensile stress to the rate of elongation) for the resulting filaments. The measurements were made on fluids of high viscosity (wax and tar) and gave values for the elongational viscosity equal to about three times the value of the shear viscosity. This was in agreement with the corresponding theoretical results for an incompressible Newtonian fluid, although it is interesting to note that Trouton observed a small recovery of the strain when the filaments were suddenly cut. Theoretically, Trouton considered the axial component of the velocity to be independent of the radial direction, i.e.,

$$V_z = V_z(z) \quad ; \quad \frac{\partial V_z}{\partial r} = \frac{\partial V_z}{\partial \theta} = 0 \tag{1.94}$$

In consequence of the continuity equation, the radial component of the velocity is given by

$$V_r = -\frac{r}{2}\frac{dV_z}{dz} \tag{1.95}$$

For an incompressible Newtonian fluid of viscosity η_o (and in the absence of surface effects) the stress distribution is given by

$$S_{rr} = -p + T_{rr} = -p - \eta_o \frac{dV_z}{dz} = 0 \tag{1.96}$$

$$S_{zz} = -p + T_{zz} = 3\eta_o \frac{dV_z}{dz} \tag{1.97}$$

For an incompressible Newtonian fluid, the elongational viscosity η_T (or, as it is sometimes called today, the <u>Trouton viscosity</u>), is given by

$$\eta_T = \frac{S_{zz}}{dV_z/dV_z} = 3\eta_o \tag{1.98}$$

The Trouton viscosity η_T is thus equal to 3 times the value the viscosity η_o, in agreement with Trouton's experimental observations.

To solve for the flow profile in the filament, Trouton set up a momentum balance obtaining

$$\frac{d(S_{zz}A)}{dz} + \rho g A + \rho A V_z \frac{dV_z}{dz} = 0 \tag{1.99}$$

where A = A(z) is the cross-sectional area of the filament and g is the acceleration of gravity. In his subsequent solution, Trouton omitted the effect of inertia (represented by the third term on the right of eq. 1.99). It is interesting to note that the sign preceding the inertia term should be negative. That is, the correct balance of momentum should read

$$\frac{d(S_{zz}A)}{dz} + \rho g A - \rho A V_z \frac{dV_z}{dz} = 0 \qquad (1.100)$$

Since Trouton's solution does not include the effect of inertia, Trouton may not have been aware of this inconsistency. Unfortunately, Trouton's work being so highly regarded, this mistake (or perhaps a misprint) found its way into the current literature. Ziabicki [75] and Pigford and Marshall [48] used the incorrect equation (1.99) in their treatment of the present problem. The correct formulation of the balance of momentum and the complete solution to eq. (1.100) are given by G.I. Taylor [62] and Clarke [10] (the former considered the analogous problem for films of fluid falling under the action of gravity). Eq. (1.100) represents also a special case of "gravitational spinning" which is considered in chapter 4.

The <u>steady extension</u> of a cylindrical filament of fluid under tension is described by the following velocity components (written in cylindrical coordinates):

$$V_z = e z \quad ; \quad V_r = -\frac{r}{2} e \quad ; \quad V_\theta = 0 \quad ; \quad e > 0 \qquad (1.101)$$

It follows from eq. (1.101) that the filament retain its circular shape at all times; it merely elongates with consequent thinning with the length L(t) and the radius R(t) of the filament given by

$$\frac{dL}{dt} = eL \quad ; \quad \frac{dR}{dt} = -\frac{e}{2}R \tag{1.102}$$

The stress distribution within the filament depends on the rheological properties of the filament fluid. Coleman and Noll [12] have obtained an exact solution for the stress pattern in a filament of an incompressible fluid which is undergoing steady extension. In the absence of surface and inertial effects the elongational viscosity η_T was found to be

$$\eta_T = \frac{S_{zz}}{e} = \frac{3}{2}q_1(e) + \frac{3}{4}q_2(e)e \tag{1.103}$$

where q_1 and q_2 are functions of the elongation rate "e". For an incompressible Newtonian fluid

$$q_1 = 2\eta_o \quad ; \quad q_2 = 0 \tag{1.104}$$

so that

$$\eta_T = 3\eta_o \tag{1.105}$$

which is identical to the result obtained by Trouton.

Experimental evidence [1, 3, 37] show that for viscoelastic fluids η_T is an <u>increasing</u> function of the rate of elongation. This is in marked contrast to the behavior of viscoelastic fluids in viscometric flows in which the shear viscosity decreases with the shear rate.

The behavior of a filament of fluid falling under the action of gravity and the steady extension of a filament of fluid under tension both represent specialized aspects of the more general problem of fiber spinning. In a typical process for the production of synthetic fibers, a molten polymer, or a solution of a polymer in a suitable solvent, is continuously extruded through narrow orifices (the spinnerette); the resulting filaments are then rapidly drawn by a wind-up device situated some distance away from the spinnerette. Molten polymers are generally extruded into an atmosphere (air) having a temperature lower than the melting range of the molten polymer, while polymer solutions are commonly drawn through a bath of liquid (or gas) into which the solvent evaporates or diffuses. The exchange of heat (or mass) with the surrounding medium results in a considerable hardening (increase in the viscosity) of the fluid filament.

A considerable literature exists on fiber spinning. A great number of these publications are the works of textile-oriented researchers whose prime interest is the textile characteristics of the finished product. Another large class of investigations deal mostly with the molecular changes which occur during the spinning process (e.g. chain folding, orientation of the molecules and crystallization). On the other hand, the number of papers which deal directly with the formation of fibers from a continuum mechanics point of view is rather small.

It has been a common knowledge for some time that fluids which exhibit elastic effects are appreciably more spinnable than fluids which are purely viscous, say Newtonian fluids. Indeed, some of the characteristic phenomena exhibited by viscoelastic fluids have been adopted as rough measures of spinnability. Among these phenomena are jet swelling and the Weissenberg climbing effect. The qualitative observation of elongated filaments drawn by hand from a polymer solution has also been taken as a proof for spinnability. The superior spinnability of viscoelastic fluids, however, is not entirely understood. At present, it appears that the dominant property of viscoelastic fluids which account for their superior spinnability is the increase of the elongational viscosity with the rate of elongation. Indeed, as early as 1948, Nitschmann and Schrade [41] postulated that this property is essential to spinning. The recent work on steady extension of viscoelastic fluids lends a substantial support to this view. However, the formation of fibers in the spinning process is a phenomenon much more complex than steady extension and direct analogies between the two are not at all obvious.

The formation of fibers depends both on the physical properties of the filament fluid and on the particular choice of spinning conditions. A rigorous mathematical description of the spinning process requires the simultaneous application of energy, mass and momentum balances. The resulting set of equations is too complex and, at present, it appears mathematically intractable. For this reason the various aspects of the spinning process are usually considered seperately. Andrews [2] , Mark [33] and Griffith [29] studied the energy transport in spinning processes involving polymer melts (melt spinning). Fok and Grisky [25] reported on mass transfer in the spinning of polymer solutions. In the present work (chapter 4) the primary interest lies in the mechanical aspects of the spinning process. That is, we wish to ascertain the effect of the rheological properties of the filament fluid upon its spinnability.

Considered even alone, the mechanical aspects of the spinning process represent a formidable analytic task. In addition to the non-linearity of the momentum equation and the constitutive equations (for non-linear viscoelastic fluids) the principal difficulty lies in the treatment of the boundary conditions at the free surface of the filament whose position is unknown. The complete problem is too difficult to solve at present and one strives to obtain a suitable approximate solution which encompasses the main features of the mechanical problem. Such an attempt has been previously made by Ziabicki and Kedzierska [75] and Ziabicki [76]. The former publication presents a simplified formulation of the momentum equation which is modeled after the work of Trouton including, unfortunately, the mistaken sign of the inertial term to which reference has been made earlier in this discussion. The subsequent discussion is, on the whole, merely qualitative and is based on the constitutive equation for a Newtonian fluid.

The latter work, by Ziabicki, represents a series of three papers in which the author attempted to obtain an approximate solution for the velocity profile in filaments of various rheological materials. A detailed appraisal of this work is beyond the scope of the present survey. Briefly, the mathematical formulation of the governing momentum equation, in this work, is based on a series of approximations which rest on numerous arbitrary assumptions which are not appropriate to the problem and for which no satisfactory justification is offered. Thus, for example, Ziabicki assumes the axial velocity in the filament to be independent of the radial direction but he errs in the treatment of the boundary conditions on the free surface of the filament. In addition, Ziabicki treats the force exerted by the wind-up roll as an external body force in the momentum equation, rather than as a quantity to be evaluated upon application of the boundary conditions at both ends of the spinning way. His analysis of the spinning of a viscoelastic material is based upon a linear constitutive model [eq. (3), p. 725] which is unrealistic and incompatible with the linear theory of viscoelasticity. Furthermore, the components of the strain appearing in the constitutive model are related to the axial velocity arbitrarily and without any apparent conformity to the momentum equation.

The spinning problem has also been treated recently by Mark [33]. However, Mark's approach is totally inadequate. The shape of the filament is obtained from the continuity equation under the assumptions that the tensile stress as well as the axial velocity are constants; the latter assumption, in particular, being unrealistic.

An elegant treatment of the spinning of an incompressible simple fluid is given by Slattery [59]. However, the mathematical description is based on a velocity profile that is assigned a priori and in the subsequent derivation of the stress distribution, surface and inertial effects are omitted.

In chapter 4 the mechanical spinning problem is formulated in its complete form. The dynamic equations are then reduced to an ordinary non-linear differential equation in the axial component of the velocity; a reduction which is valid for the case in which the rate of elongation is small.

Solutions to the reduced problem are obtained corresponding to the spinning of filaments of Newtonian and non-Newtonian fluids.

CHAPTER 2

THE RESPONSE OF VISCOELASTIC MATERIALS TO SMALL DEFORMATIONS

2.1 THE RESPONSE OF VISCOELASTIC MATERIALS TO SMALL DEFORMATIONS NEAR EQUILIBRIUM

We commence our discussion by postulating the existence of a linear stress-strain relationship for a viscoelastic fluid which is subjected to a sufficiently small deformation. Thus,

$$\underset{\sim}{S} = L(\underset{\sim}{E}^{(L)}) \tag{2.1}$$

where $\underset{\sim}{S}$ and $\underset{\sim}{E}^{(L)}$ are the stress and the linear strain tensors respectively. The properties of the linear operator L and its functional form should be deduced from general principles of continuum mechanics without resorting to postulates which can not be assumed a priori. While the mechanical-model approach to linear viscoelasticity has led to useful results, it does not represents a well-founded theory and therefore admits doubts as to the validity of the results in general. An elegant theoretical approach which enjoys a sufficient generality has been adopted by Biot [5]. Biot formulates the behavior of a viscoelastic material as a relaxation process governed by a number of "hidden" thermodynamic variables. The behavior of the material is characterized by two quadratic forms, the potential energy (which appears in the construction of the entropy function) and the dissipation function (which gives the entropy production rate). Application of Onsager's reciprocal relations† then leads to a form of constitutive equation which can represent the behavior of any viscoelastic material in the vicinity of equilibrium. Linearity is imposed by assuming the state variables to be linear functions of the driving forces.

Bland [6], in a different approach, develops the properties of a linear viscoelastic material from the hypothesis that the microscopic structure of such a material is mechanically equivalent to a network of elastic and viscous elements. Linearity is ensured by assuming that the relative rotations of the elements are small. Relations between the material constants are based on thermodynamic considerations.

For an isotropic material, the independent analyses of Biot and Bland both show that; a) each deviatoric component of the stress is solely related to the corresponding component of the strain; the dilatational part of the stress is only related to the dilataional part of the strain; b) the stored energy and the dissipation are the sum of the respective energies of the deviatoric components and the dilatation. The time-dependent relationship between any component of the stress and the corresponding component of the strain can then be described by

$$T(t) = h\,E^{(L)}(t) + \eta_{\infty}\frac{dE^{(L)}}{dt}(t) + \sum_{r=1}^{N}\int_{-\infty}^{t} C_r\, e^{-\mu_r(t-\tau)}\, dE^{(L)}(\tau) \qquad (2.2)$$

† Some of the aspects of the thermodynamic theory which places restrictions on the constitutive equations of linear viscoelasticity have also been deduced without the application of the, often criticized, Onsager's reciprocal relations [13, 69].

where h, η_∞, C_r and μ_r are all real and positive. The coefficients h and η_∞ account for the instantaneous elasticity and long term viscous flow respectively. The summation represents a discrete spectrum of relaxation times $1/\mu_r$. Corresponding to eq. (2.2) is a complex viscosity $\hat{\eta}(\alpha)$ obtained via a two-sided Laplace transform and defined as

$$\hat{\eta}(\alpha) = \frac{\hat{T}(\alpha)}{\alpha \hat{E}^{(L)}(\alpha)} = \frac{h}{\alpha} + \eta_\infty + \sum_{r=1}^{N} \frac{C_r}{\mu_r + \alpha} \tag{2.3}$$

The constitutive equation (2.2) thus represents the form of the general linear relationship of eq. (2.1) dictated by considerations of irreversible thermodynamics or, alternatively, by the behavior of networks of springs and dash-pots. The finite number N of the relaxation terms appearing in eq. (2.2) corresponds on the one hand to the finite number of hidden variables in the thermodynamic relaxation process, and on the other, to the finite number of springs and dash-pots used to model the material. Since real materials are not necessarily bound by this finiteness, eq. (2.2) should perhaps be regarded as a family of stable approximations to the linear behavior of real materials.

The complex viscosity $\hat{\eta}(\alpha)$ of eq. (2.3) serves as a compact description of the constitutive equation (2.2). It is, in fact, simply the stress-strain rate transfer function for the material, and the behavior of the material can be described in terms of the analytic character of $\hat{\eta}(\alpha)$, considered as a function of the complex variable α. Thus, the $\hat{\eta}(\alpha)$ of eq. (2.3) has simple poles at $\alpha = 0$ (corresponding to the purely elastic response), and at values $\alpha = -\mu_r$ (corresponding to the relaxation times). The residues at these poles are all positive, and further, $\hat{\eta}(\alpha)$ approaches the positive value η_∞ as $\alpha \to \infty$, and (when h = 0) the positive value $\eta_0 = \eta_\infty + \sum_{r=0}^{N} C_r/\mu_r$ as $\alpha \longrightarrow 0$. As in eq. (2.2), this simple character corresponds to the finiteness of N, and eq. (2.3) should also be regarded as a family of stable (rational) approximations to the complex viscosities of real materials.

2.2 ON LINEAR EXPERIMENTS

Experimental studies in linear viscoelasticity are generally reported in terms of stress-strain or stress-strain rate ratios which are obtained under an appropriate time-dependent pattern; the constitutive equations can then be fitted to these measured values. In principle, linear experiments should yield the same information regardless of the loading pattern. By far, the most popular experimental technique is the one involving the application of a sinusoidally varying stress to a viscoelastic body. The viscoelastic properties of such a body can then be completely determined from measurements of the complex modulus (stress-strain ratio), the complex compliance (strain-stress ratio) or the complex viscosity as a function of the frequency.

One of the consequences of the linear theory of viscoelasticity, as outlined in the previous section, is the requirement that the absolute value of the complex viscosity, as well as its real part, be monotonically decreasing functions of the frequency. To prove this assertion, we first break the complex viscosity into its real and imaginary parts. From eq. (2.3), with α replaced by iw,

$$\hat{\eta}(iw) = \eta' - \eta'' = \left[\eta_\infty + \sum_{r=1}^{N} \frac{C_r \mu_r}{\mu_r^2 + w^2}\right] - i\left[\frac{h}{w} + \sum_{r=1}^{N} \frac{C_r w}{\mu_r^2 + w^2}\right] \tag{2.4}$$

It is clear from eq. (2.4) that the real part, η', of the complex viscosity is a decreasing function of w. To verify that the absolute value is as well, we form its square

$$|\hat{\eta}|^2 = (\eta')^2 + (\eta'')^2 = \left[\eta_\infty + \sum_{r=1}^{N} \frac{C_r \mu_r}{\mu_r^2 + w^2}\right]^2 + \left[\frac{h}{w} + \sum_{r=1}^{N} \frac{C_r w}{\mu_r^2 + w^2}\right]^2 \tag{2.5}$$

or

$$|\hat{\eta}|^2 = \eta_\infty^2 + 2\eta_\infty \sum_{r=1}^{N} \frac{C_r \mu_r}{\mu_r^2 + w^2} + \sum_{r,s=1}^{N} \frac{C_r \mu_r}{\mu_r^2 + w^2} \frac{C_s \mu_s}{\mu_s^2 + w^2} + \frac{h^2}{w^2}$$

$$+ \frac{2h}{w} \sum_{r=1}^{N} \frac{C_r w}{\mu_r^2 + w^2} + \sum_{r,s=1}^{N} \frac{C_r w}{\mu_r^2 + w^2} \frac{C_s w}{\mu_s^2 + w^2} \tag{2.6a}$$

$$|\hat{\eta}|^2 = \eta_\infty^2 + \frac{h^2}{\omega^2} + 2\eta_\infty \sum_{r=1}^{N} \frac{C_r \mu_r}{\mu_r^2 + \omega^2} + 2h \sum_{r=1}^{N} \frac{C_r}{\mu_r^2 + \omega^2}$$

$$+ \sum_{r,s=1}^{N} \frac{C_r C_s (\mu_r \mu_s + \omega^2)}{(\mu_r^2 + \omega^2)(\mu_s^2 + \omega^2)} \tag{2.6b}$$

Examining eq. (2.6), it is obvious that, with the possible exception of the terms in the double sum on the right, all other terms decrease monotonically with w. To show that a typical term in the last summation behaves similarly, we differentiate it with respect to w,

$$\frac{d}{d\omega}\left[\frac{C_r C_s (\mu_r \mu_s + \omega^2)}{(\mu_r^2 + \omega^2)(\mu_s^2 + \omega^2)}\right] = \frac{2C_r C_s \mu_r \mu_s [\mu_r \mu_s - \mu_r^2 - \mu_s^2 - 2\omega^2 - \omega^4/(\mu_r \mu_s)]}{(\mu_r^2 + \omega^2)^2 (\mu_s^2 + \omega^2)^2}$$

and note that

$$\mu_r \mu_s < \mu_r^2 + \mu_s^2$$

We have thus shown that, according to the linear theory, the absolute value of the complex viscosity is a monotonically decreasing function of the frequency. We shall later make use of this property in evaluating the relative stability of a viscoelastic filament with respect to either a Newtonian or purely elastic filaments.

Experimental evidence generally confirm the frequency-dependent behavior of the complex viscosity as predicted by the linear theory. Some experimental data reported on in the literature shows slight deviations from this behavior, but these could very well be within the range of the experimental error. It is suggested, nonetheless, that the requirement that $|\hat{\eta}|$ be a decreasing function of the frequency be used as an additional test in checking the linearity of experimental results.

Linear viscoelastic measurements suffer from certain limitations which are inherent in the experimental instrumentation. The proper design of a suitable test apparatus is very complex, particularly when information is sought in the lower end of the frequency range where a small signal-to-noise ratio may result in misleading data. In linear experiments, several criteria are used to ensure the linearity of the data. In vibration experiments one such criterion is the requirement that $\hat{\eta}(iw)$ be independent of the amplitude of the driving force. This amplitude test is normally applied at a few selected frequencies. Now, the standard instruments do not record the total response over all frequencies, but rather, the amplitude and the phase angles corresponding to specific test frequencies. Such filtering is often necessary due to the noise generated in the system. This procedure, therefore, does not necessarily ensure that the system is linear. To illustrate this, consider the following hypothetical example: Suppose that a given material is described by:

$$S(t) = \begin{cases} \eta_\infty \dfrac{dE(t)}{dt} + hE(t) & ; \quad |E| < E_{cr} \\ \\ \eta_\infty \dfrac{dE(t)}{dt} + hE_{cr}\,\mathrm{sgn}.\,E(t) & ; \quad |E| \geqslant E_{cr} \end{cases} \tag{2.7}$$

This is a Voigt material below the critical strain E_{cr}, and a Bingham fluid above.

Let

$$\lambda = \eta_\infty/E \quad , \; S_o = hE_{cr} = \eta_\infty E_{cr}/\lambda$$

and rewrite eq. (2.7) in the form

$$S(t) = \eta_\infty \frac{dE(t)}{dt} + f\left[E(t)\right] \tag{2.8}$$

where

$$f\left[E(t)\right] = \begin{cases} \eta_\infty E(t)/\lambda & ; \quad |E| < \lambda S_o/\eta_\infty \\ \\ S_o\,\mathrm{sgn}.\,E(t) & ; \quad |E| \geqslant \lambda S_o/\eta_\infty \end{cases}$$

Consider now a sinusoidal strain of frequency w applied to the material,

$$E(t) = K^\circ \cos \omega t \quad ; \quad K^\circ, \omega > 0$$

The resulting periodic stress (after initial transients have died out) is given by the Fourier series

$$S(t) = \frac{a_o}{2} + \sum_{n=1}^{\infty} a_n \cos n\omega t + \sum_{n=1}^{\infty} b_n \sin n\omega t \tag{2.9}$$

where

$$a_n = \frac{\omega}{\pi}\int_{-\pi/\omega}^{\pi/\omega} f\left[E(t)\right]\cos n\omega t\,dt \quad ; \quad b_n = \frac{\omega}{\pi}\int_{-\pi/\omega}^{\pi/\omega} f\left[E(t)\right]\sin n\omega t\,dt$$

We may note that

$$\text{sgn}\cdot E(t) = \begin{cases} +1 & ; \quad |t| < \frac{\pi}{2w} \\ -1 & ; \quad \frac{\pi}{2w} < |t| < \frac{\pi}{w} \end{cases}$$

When $K^{o} < \lambda S_o/\eta_\infty$ the material exhibits a linear behavior, namely, that of a simple Voigt solid,

$$S(t) = -\eta_\infty K^{o}\left[\omega \sin \omega t - \frac{1}{\lambda}\cos \omega t\right] \tag{2.10}$$

with complex viscosity

$$\hat{\eta}(i\omega) = \eta_\infty\left[1 - \frac{i}{\lambda w}\right] \tag{2.11}$$

On the other hand, consider the case for which $K^{o} >> \lambda S_o/\eta_\infty$. Eq. (2.9) then reduces to

$$S(t) = -\eta_\infty K^{o} w \sin \omega t + \frac{4S_o}{\pi}\cos \omega t - \frac{4S_o}{3\pi}\cos 3\omega t$$

$$+ \frac{4S_o}{5\pi}\cos 5\omega t - \cdots \tag{2.12}$$

At the fundamental frequency w, the response is

$$S(t) \cong -K^{o} w\left[\eta_\infty \sin \omega t - \frac{4S_o}{K^{o}\pi w}\cos \omega t\right]$$

$$= g(w) K^{o} w \sin\left[wt - \phi(w)\right] \tag{2.13}$$

where

$$g(w) = \eta_\infty\left[\eta_\infty^2 + \left(\frac{4S_o}{K^{o}\pi w}\right)^2\right]^{-1/2}; \quad \phi(w) = \frac{4S_o}{\eta_\infty K^{o}\pi w}$$

so that the complex viscosity is

$$\hat{\eta}(i\omega) = g(\omega)\, e^{-i\phi(\omega)} = \eta_\infty - i\,\frac{4S_0}{k^\circ \pi \omega} \tag{2.14}$$

One might accordingly by pre-filtering conclude that the material is linear and purely viscous, having entirely overlooked the harmonics which are not small. The above example demonstrates, therefore, that special care should be taken to ensure that the experimental results are truly linear.

2.3 SMALL PERTURBATIONS SUPERPOSED UPON STEADY SHEAR FLOW

The response of viscoelastic materials to small perturbations superimposed on a finite steady deformation is of great interest in many applications. Knowledge of such a response could, for example, be utilized in linear analyses of the stability of viscoelastic bodies that are undergoing steady deformation, relative to small disturbances or, in the study of wave propagation on the surface of initially stressed bodies. Several experimental works pertaining to the above problem have been published in the past few years. The corresponding theoretical problem, however, has received little attention and the few published reports deal mostly with viscoelastic solids. Our purpose in this section is to examine some of the concepts which have been formed as a result of the studies dealing with viscoelastic fluids. To this end we restrict ourselves to one specific representative problem namely, the response of viscoelastic fluids to small sinusoidal shearing motion superimposed upon some primary steady shear flow. In fact, for viscoelastic fluids the only problem which has been considered so far is their response to small perturbations superposed on a steady shear flow.

Experimental results [8, 45, 58, 61] show that, in the presence of steady shear, the dynamic response of viscoelastic fluids to small sinusoidal motion is highly dependent on the steady shear rate. This dependence is manifested by the measured values of the complex viscosity corresponding to the oscillating components of the stress and the shear rate. In general, the values of the real and the imaginary parts of the complex viscosity decrease strongly with increasing rate of steady shear. The effect of the steady shear on the complex viscosity of the oscillating components have generally been attributed, by the investigators cited above, to the notion that the steady shearing motion obliterates the long relaxation times so that the response of the sheared fluid is "less elastic" than one might expect from a spectrum measured in a fluid with zero mean shear rate.

Such a notion is based on the assumption that the dynamic response of viscoelastic fluids to small deformations is governed by the same physical principles regardless of whether the fluid is initially at rest or is undergoing steady shear flow. However, small perturbations about a quiescent state constitute small deviations from an equilibrium state, and as such are governed by thermodynamical considerations which, in general, are not applicable to perturbations about a finite steady flow. Thus, for example, while any true thermodynamic equilibrium is, by definition, stable relative to infinitesimal perturbations, steady motions are of course not necessarily stable with respect to such perturbations. This is not to say that the respective dynamic responses of a given fluid in the presence of steady shear and near equilibrium may not show some similar properties; however, because of their fundamental differences, the two types of response can not be a priori interpreted on the same physical basis.

It was shown in the previous section that the dynamic behavior of viscoelastic materials near equilibrium is characterized by a complex viscosity $\hat{\eta}(\alpha)$ having simple poles which are situated along the negative real axis, with corresponding residues all of which are real and positive. Consequently, the real part and the absolute value of the complex viscosity $\hat{\eta}(iw)$, measured in vibrational experiments, are decreasing functions of the frequency. In the case of fluids under steady shear, if the effect of the steady shear is merely to eliminate the long relaxation times, the the real part and the absolute value of the complex viscosity of the overlying sinusoidal motion should also be decreasing functions of the frequency. Turning to the experimental evidence, the complex viscosities measured by Booij [8] and Osaki, et al. [45], indeed exhibit this type of behavior. On the other hand, the complex viscosities obtained by Tanner and Simmons [61, 58], show significant departures from this type of frequency-dependent behavior. Figs. 2.1, 2.2 and 2.3 reproduce some of their experimental results. Figs. 2.1 and 2.3 show that, in the presence of steady shear, the real part and the absolute value of the complex viscosities both display a maximum with respect to the frequency; the effect is more pronounced with increasing values of the steady shear rate. The presence of a maximum in the viscosity curves is not compatible with the infinitesimal theory for perturbations about equilibrium. Thus, if we compare the above results to those obtained for a fluid near equilibrium (K = 0) on an equivalent basis, we would have to conclude that the results of Tanner and Simmons do not represent a proper linear response. The amplitude of the perturbations may have been too large leading to a non-linear response in the sense that the superposition principle no longer holds and one can not isolate the response of the fluid to the extra motion .

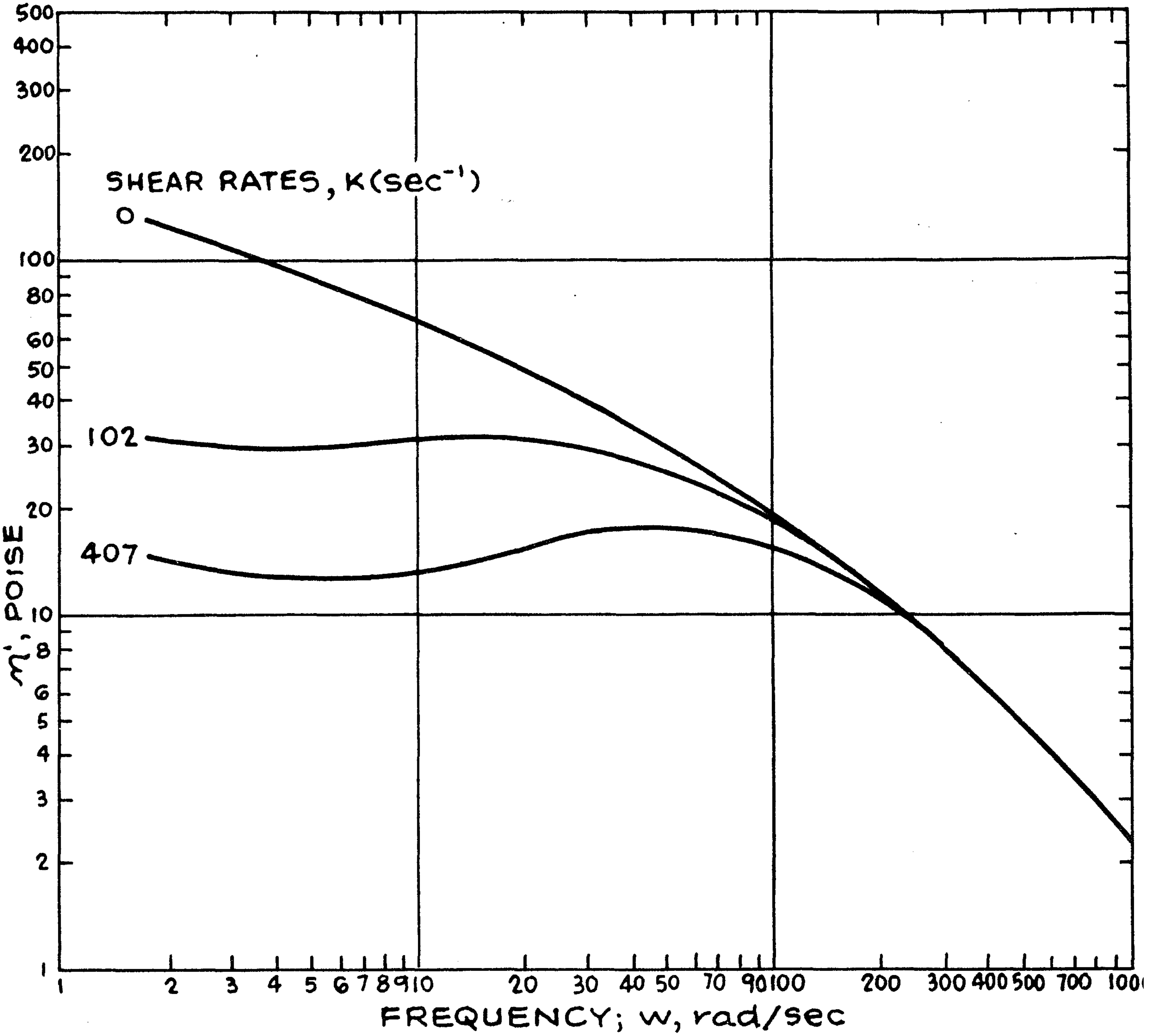

FIG. 2.1 THE REAL PART, η', OF THE COMPLEX VISCOSITY OF 8.7% POLYISOBUTYLENE-CETANE SOLUTION (AT 25 °C) WITH AND WITHOUT SHEARING [58,61].

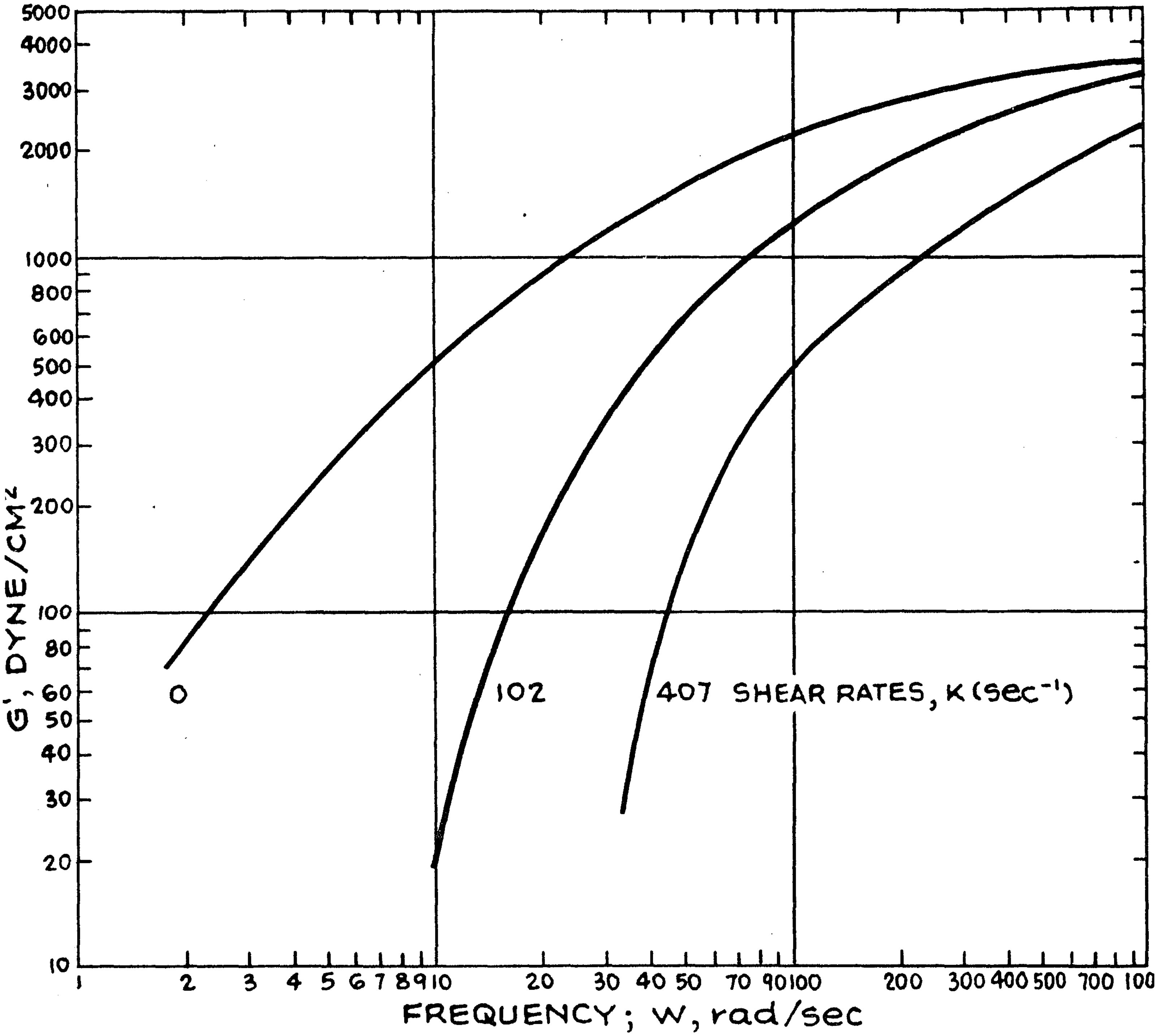

FIG. 2.2 THE REAL PART OF THE COMPLEX MODULUS ($G' = \eta'' W$) OF 8.7% POLYISOBUTYLENE-CETANE SOLUTION (AT 25°C) WITH AND WITHOUT SHEARING [58,61].

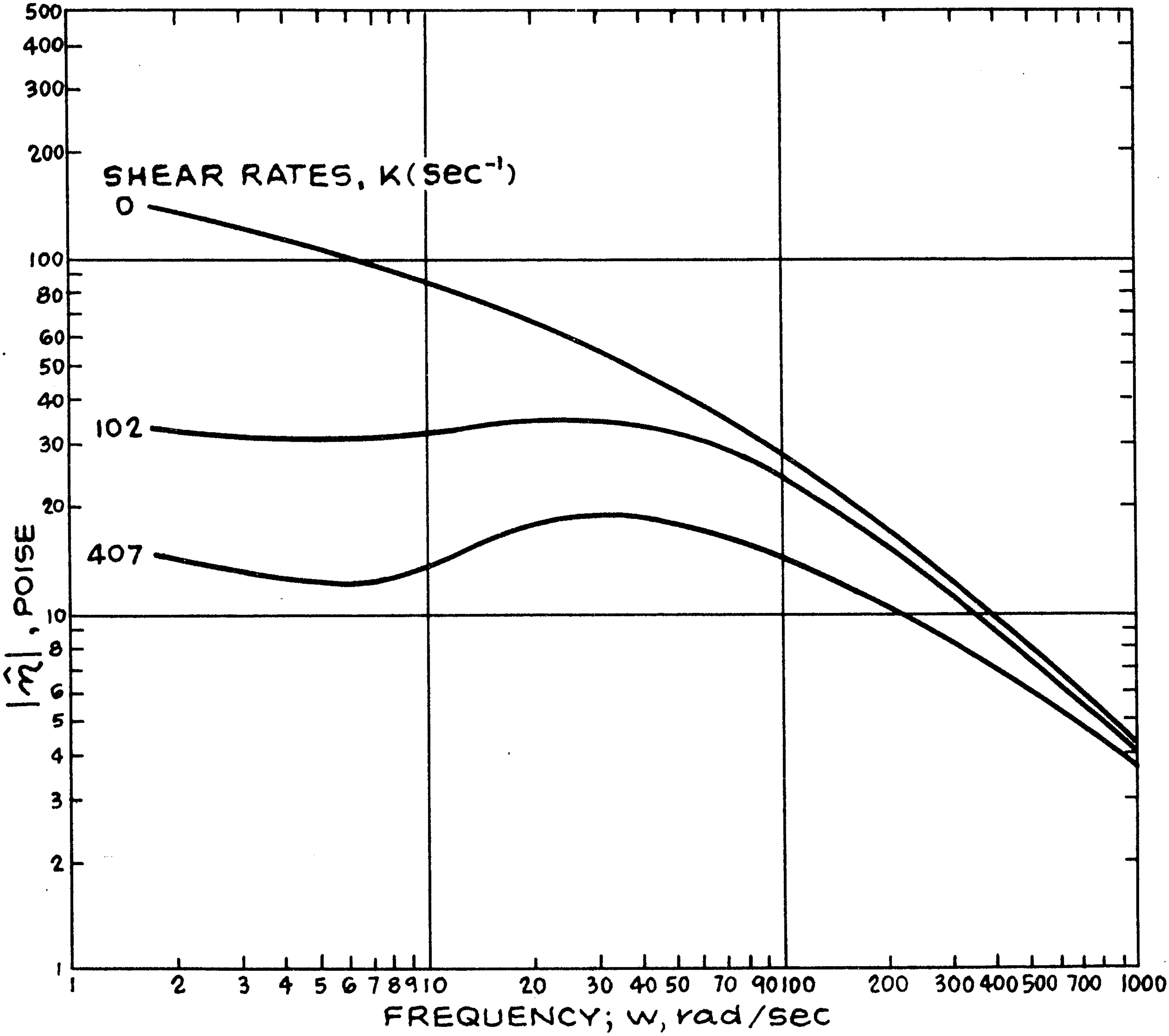

FIG. 2.3 THE ABSOLUTE VALUE OF THE COMPLEX VISCOSITY OF 8.7% POLYISOBUTYLENE-CETANE SOLUTION (AT 25°C) WITH AND WITHOUT SHEARING [58, 61].

Finite perturbation imposed on a strongly sheared fluid may also cause secondary flows which would naturally affect the complex viscosity. These conclusions, however, are not necessarily true. It is equally possible that the results of Tanner and Simmons are linear and do indeed represent the dynamic behavior of viscoelastic fluids under shear. As was pointed out before, the behavior under steady shear is not bound by the same constraints which apply to small deformations near equilibrium. It is thus possible that, in the presence of steady shear, the poles of the complex viscosity $\hat{\eta}(\alpha,K)$ (where the argument K emphasizes the dependence on the primary steady shear) are no longer confined to the negative real axis, but may lie anywhere in the left half of the complex plane. Under such circumstances, the real part and the absolute value of the complex viscosity $\hat{\eta}(iw,K)$ need not be decreasing functions of the frequency, and may very well exhibit a maximum. Several theoretical models, commonly used by rheologists, predict that, in the presence of steady shear, the singularities of $\hat{\eta}(\alpha,K)$ may indeed become complex. Booij [8], considered the following non-linear constitutive equation

$$\underset{\sim}{T} + \lambda \frac{\mathcal{D}\underset{\sim}{T}}{\mathcal{D}t} = 2\eta_0 \underset{\sim}{D} \qquad (2.15)$$

Eq. (2.15) represents one of the non-linear generalizations of the linear Maxwell fluid discussed earlier.

Let D_{12}, the only non-vanishing component of the rate of strain tensor, be given by

$$D_{12} = \frac{1}{2}\left[K + Re\,(K^{o} e^{i\omega t}) \right] \tag{2.16}$$

and postulate that the induced stresses are of the form

$$T_{ij}^{o} = \bar{T}_{ij} + Re\,(K^{o} e^{i\omega t}) \tag{2.17}$$

where T_{ij}^{o} are the complex amplitudes of the stresses generated by the oscillating motion, and $\bar{T}_{ij}$ their mean value. In the limit of small K^{o}, the complex viscosity $\hat{\eta}(iw,K)$, corresponding to the superposed motion, can then be obtained by a straight forward perturbation analysis. Accordingly,

$$\frac{T_{12}^{o}}{K^{o}} = \hat{\eta}(i\omega, K) = \frac{\eta_o}{(1+\lambda^2 K^2)} \frac{[1 - \lambda^2 K^2 + i\omega\lambda]}{[(1 + i\omega\lambda)^2 + \lambda^2 K^2]} \tag{2.18}$$

and $\hat{\eta}(\alpha,K)$ is

$$\hat{\eta}(\alpha, K) = \frac{\eta_o}{(1+\lambda^2 K^2)} \frac{[1 - \lambda^2 K^2 + \lambda\alpha]}{[1 + \lambda^2 K^2 + 2\lambda\alpha + \lambda^2\alpha^2]} \tag{2.19}$$

Eq. (2.19) shows that $\hat{\eta}(\alpha,K)$ has two simple poles situated at

$$\alpha = -1/\lambda \quad \pm iK \tag{2.20}$$

Thus, for any finite K, the values of α in eq. (2.20)(corresponding to the poles of $\hat{\eta}(\alpha,K)$) are both <u>complex</u>. Fig. 2.4

shows that the real part of the complex viscosity of eq.(2.18) indeed exhibit a maximum.

Similar results have been obtained by the present author through the use of other common constitutive equations of the rate as well as the integral type. Note that in eq. (2.20), the real part of α is identical to the value of α given by the infinitesimal theory, while the imaginary part is proportional to the velocity of the steady flow. One might hence conclude that the effect of the steady shear is <u>not</u> to eliminate any of the relaxation times, but rather, to shift the position of the poles of $\hat{\eta}(\alpha,K)$ away from the negative real axis by an amount proportional to the value of the steady shear rate. Whether or not this is a general property of real fluids can not be established with any certainty since, at present, our knowledge of proper constitutive equations for real fluids is quite incomplete†. However, if the results of Tanner and Simmons are indeed linear, these results

† Pipkin and Owen [49], have developed a general theory for small deformations superposed on steady shear flows. The resulting equations are linear in the history of the perturbed deformation and resemble the integral stress-strain relation of linear viscoelasticity. The memory functions appearing in this integral theory are functions of time as well as the basic invariants of the primary motion. This theory, however, yields no specific information on the properties of the memory functions and as such can provide no further insight to the above discussion.

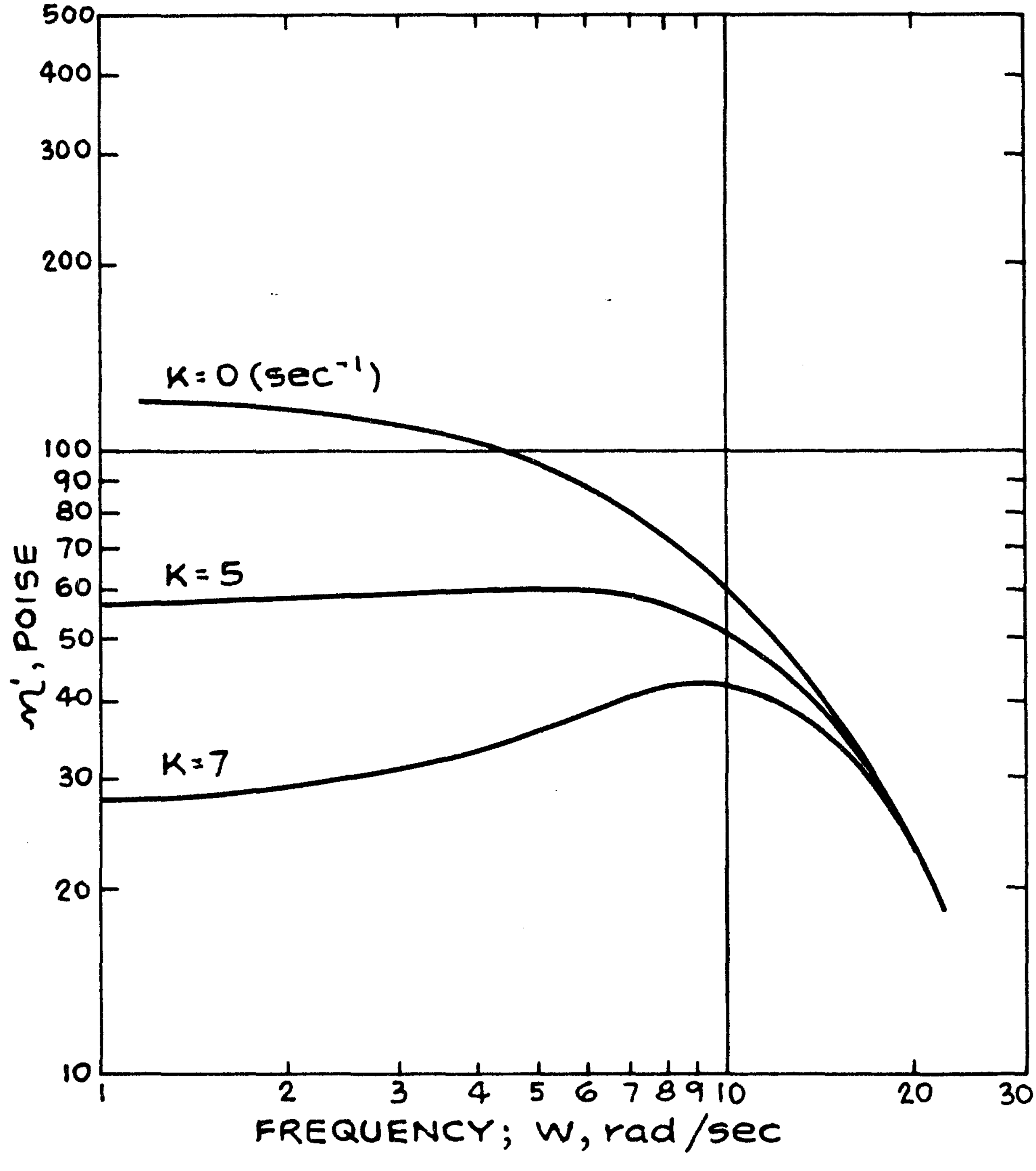

FIG. 2.4 THE REAL PART, η', OF THE COMPLEX VISCOSITY GIVEN BY EQ.(2.18). η_0 = 120 POISE, λ = 0.1 SEC.

would seem to corroborate the above conclusion. In any event, the experimental and theoretical evidence presented here should constitute a strong indication of the essential differences between the properties of the complex viscosities representing the dynamic response of viscoelastic fluids in the presence of steady motions and in the vicinity of equilibrium.

2.4 CONSEQUENCES OF THE LINEAR THEORY FOR NON-LINEAR VISCOELASTICITY

The linear theory of viscoelasticity describes the behavior of viscoelastic materials in the region of very small deformations. It is obvious that the linear theory ceases to be adequate as soon as these conditions are not fulfilled. Many of the properties of viscoelastic materials are inherently non-linear (e.g. non-linear viscosity in viscometric flows). To describe the non-linear behavior of viscoelastic materials, a suitable non-linear theory in needed. Several of the more common non-linear constitutive theories have been reviewed in the previous chapter; some of the principles which guide the formulation of an appropriate theory have also been discussed. Here we wish to examine one of the consequences of the linear theory for non-linear viscoelasticity. Since the validity of the linear theory is universally acknowledged, we require that any non-linear theory be compatible with the linear theory of viscoelasticity. That is, when linearized about a quiescent state, any non-linear constitutive equation should reduce to an equation whose time-dependent behavior is equivalent to that predicted by the linear theory.

To illustrate the consequences of this criterion, consider a class of non-linear constitutive equations of the form

$$P(\underset{\sim}{T}) = Q(\underset{\sim}{D}) \qquad (2.21)$$

where P and Q are non-linear operators involving time derivatives. In the linear region of deformations, eq. (2.21) gives rise to a linear equation with a corresponding complex viscosity given by

$$\hat{\eta}(\alpha) = \frac{\hat{Q}_L(\alpha)}{\hat{P}_L(\alpha)} \qquad (2.22)$$

where P_L and Q_L are the linearized counterparts of the non-linear operators P and Q. To be compatible with the linear theory, the complex viscosity of eq. (2.22) should either be of the form of eq. (2.3) or allow a suitable approximation in this form. We recapitulate here, for convenience, the analytic properties noted earlier of the complex viscosity of the form of eq. (2.3) with h = 0. These are rational functions of α with the following properties:

a) $\hat{\eta}(0) > 0$

b) $\lim\limits_{\alpha \to \infty} \hat{\eta}(\alpha) > 0$

c) The singularities of $\hat{\eta}(\alpha)$ are simple poles all lying on the negative real axis.

d) Corresponding to the poles, the residues are all real and positive.

In many instances the operators P and Q of eq. (2.21) are approximated by polynomials; consequently, the complex viscosity corresponding to the linearized equation becomes a ratio of polynomials in α with constant coefficients,

$$\hat{\eta}(\alpha) = \frac{\hat{Q}_L(\alpha)}{\hat{P}_L(\alpha)} = \frac{\sum\limits_{n=0}^{N} q_n \alpha^n}{\sum\limits_{n=0}^{N} p_n \alpha^n} \tag{2.23}$$

Since the $\hat{\eta}(\alpha)$ of eq. (2.23) is already a rational function, no question of further approximation arises, and conformity to the linear theory requires simply that the conditions a, b, c and d above be satisfied. This can be established specifically in terms of the properties of the polynomials $\hat{P}_L$ and $\hat{Q}_L$. By using the theory of partial fractions Bland has shown that eq. (2.23) is identical to eq. (2.3) (with h = 0) if the zeros of $\hat{P}_L(\alpha)$ and $\hat{Q}_L(\alpha)$ are all real and non-positive and they alternate, the least zero in absolute magnitude belonging to $\hat{Q}_L(\alpha)$. The polynomials $\hat{P}_L(\alpha)$ and $\hat{Q}_L(\alpha)$ must of course be of the same degree. Thus, eq. (2.23) can be written as

$$\hat{\eta}(\alpha) = \eta_\infty \frac{\prod_{b=1}^{N} (\alpha + \lambda_b)}{\prod_{r=1}^{N} (\alpha + \mu_r)} \tag{2.24}$$

where the $-\lambda_b$ are the zeros of $\hat{Q}_L(\alpha)$, and correspond to a discrete spectrum of retardation times $1/\lambda_b$.

If for any real fluid $\eta_\infty = 0$, eq. (2.24) should then be modified to read

$$\hat{\eta}(\alpha) = \gamma \frac{\prod_{b=1}^{N} (\alpha + \lambda_b)}{\prod_{r=1}^{N+1} (\alpha + \mu_r)} \quad ; \; \gamma > 0 \tag{2.25}$$

with the special case of

$$\hat{\eta}(\alpha) = \frac{C}{\alpha + \mu} \tag{2.26}$$

representing the complex viscosity of a simple Maxwell fluid.

Turning to some specific examples, consider first the constitutive equation for a three-constants Oldroyd fluid, given by

$$\underset{\sim}{T} + \lambda_1 \frac{\mathcal{D}\underset{\sim}{T}}{\mathcal{D}t} = 2\eta_\infty \left[\underset{\sim}{D} + \lambda_2 \frac{\mathcal{D}\underset{\sim}{D}}{\mathcal{D}t} \right] \tag{2.27}$$

The complex viscosity of the linearized equation is

$$\hat{\eta}(\alpha) = \eta_0 \frac{1 + \lambda_2 \alpha}{1 + \lambda_1 \alpha} = \eta_0 \left[\frac{\lambda_2}{\lambda_1} + \frac{(\lambda_1 - \lambda_2)/\lambda_1}{1 + \lambda_1 \alpha} \right] \tag{2.28}$$

In consequence of the restrictions on $\hat{\eta}(\alpha)$ discussed above, the fluid constants λ_1 and λ_2 must obey

$$\lambda_1 > 0 \quad ; \quad \lambda_2 > 0 \quad ; \quad \lambda_1 > \lambda_2 \tag{2.29}$$

On the other hand consider the non-linear constitutive equation of Rivlin and Ericksen which reads

$$\underset{\sim}{T} = \underset{\sim}{F}\left[\underset{\sim}{A}^{(1)}, \underset{\sim}{A}^{(2)} \ldots \underset{\sim}{A}^{(N)}\right] \tag{2.30}$$

where $\underset{\sim}{A}^{(N)}$ is the Nth Rivlin-Ericksen kinematic tensor. The complex viscosity corresponding to the linearized equation is

$$\hat{\eta}(\alpha) = \sum_{n=0}^{N} a_n \alpha^n \tag{2.31}$$

The Rivlin-Ericksen polynomial of eq. (2.30) contains of course the case of a Newtonian fluid (degree N = 0, $a_0 > 0$). But for no higher-order polynomial is eq. (2.30) compatible with the linear theory as represented by the complex viscosity of eq. (2.3) or of eq. (2.24). Such polynomials simply do not have the analytic properties noted earlier; for example, they do not have the correct behavior in the limit of large α. This is not of course to say that a suitable power series in α can not serve as complex viscosities. Thus

$$\hat{\eta}(\alpha) = \eta_\infty + \frac{C}{\mu} + \frac{C}{\mu}\sum_{n=0}^{\infty}\left(\frac{-1}{\mu}\right)^n \alpha^n \tag{2.32}$$

is simply another way of writing

$$\hat{\eta}(\alpha) = \eta_\infty + \frac{C}{\mu + \alpha} \tag{2.33}$$

which is the form of eq. (2.3) with a single relaxation time (and h = 0). Consequently, a Rivlin-Ericksen equation which linearizes to eq. (2.32) with η_∞, C and μ all > 0, is compatible with the linear theory. On the other hand, any finite number of terms of eq. (2.32), say

$$\hat{\eta}(\alpha) = \eta_\infty + \frac{C}{\mu} - \frac{C}{\mu^2}\alpha \qquad (2.34)$$

while approximating the behavior of eq. (2.32) for small α no longer has the same general analytic character. That is to say, equations of the Rivlin-Ericksen type of finite degree N greater than zero should not be regarded as proper constitutive equations. Note that eq. (2.34) is equivalent to the complex viscosity corresponding to the linearized equation of the second-order fluid. It may be recalled that the constitutive equation for a second-order fluid represents a second-order approximation to the behavior of an incompressible simple fluid in slow flows; the first order approximation being the theory of Newtonian fluids. Now, the Newtonian theory is generally regarded, not as approximate, but rather as an exact, self-contained theory describing the behavior of a large class of real fluids over an unrestricted range of flow conditions. Obviously such is not the case with the second-order theory. Granting its status as an approximate theory for slow flows, in general it can not be regarded as an appropriate theory.

Finally, it may be noted that a proper constitutive equation of the form of eq. (2.30) involving derivatives of all orders (infinite N) does not really gives the stress as a differential operator on the strain. Indeed, the infinite sequence of derivatives appearing in eq. (2.30) might perhaps better be regarded as an abstract extension of what are essentially integral operators on the strain rate, whose linearization is of the form of eq. (2.2).

CHAPTER 3

THE STABILITY OF A LAMINAR CAPILLARY JET OF A VISCOELASTIC FLUID

3.1 SOLUTION OF THE DYNAMIC EQUATIONS

Consider a cylindrical jet issuing from a nozzle of radius R_o into air. The following assumptions are made:

1) The jet is initially relaxed moving horizontally with a uniform velocity of V_o.
2) The capillary waves are symmetrical about the jet axis; that is, the jet is at all times circular in cross section and either expands or contracts.
3) The fluid is incompressible.
4) There is no interaction with the ambient air.
5) Perturbations of the jet surface as well as the perturbed velocities are infinitesimally small.

Because of the symmetry of the problem, a cylindrical coordinate system (r, θ , z) is chosen which moves horizontally with the jet at a constant velocity of V_o. It follows from assumption 5) that terms proportional to $\underset{\sim}{v}\cdot\nabla\underset{\sim}{v}$ appearing in the equations of motion are of second order of smallness and, hence, can be neglected. With the usual notation, the linearized equations of motion and continuity are,

$$\rho \frac{\partial V_r}{\partial t} = - \frac{\partial p}{\partial r} + \frac{1}{r}\frac{\partial}{\partial r}(r\, T_{rr}) + \frac{\partial T_{zz}}{\partial z} \qquad (3.1a)$$

$$\rho \frac{\partial V_z}{\partial t} = - \frac{\partial p}{\partial z} + \frac{1}{r}\frac{\partial (r T_{rz})}{\partial r} + \frac{\partial T_{zz}}{\partial z} \tag{3.1b}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r V_r) + \frac{\partial V_z}{\partial z} = 0 \tag{3.2}$$

Throughout this development, the components of $\underset{\sim}{v}$ and $\underset{\sim}{T}$ represent perturbed quantities only; hence we omit our convention of designating perturbed quantitities by the superscript (^{o}). The position of the free surface is given by

$$R_o + \xi \tag{3.3}$$

where $\xi = \xi(z,t)$ is a small radial displacement of a point on the surface of the jet, and it is assumed that

$$\xi \ll R_o \tag{3.4}$$

The boundary conditions specify that the shear stress vanishes on the surface of the jet and that the radial component of the stress is balanced by the stress induced by the surface tension force. It follows from eq. (3.4) that the boundary conditions can be expressed as

$$\begin{aligned} \left[T_{rz}\right]_{r \sim R_o} &= 0 \\ \left[-p + T_{rr}\right]_{r \sim R_o} &= 2H\sigma \end{aligned} \tag{3.5}$$

where σ is the coefficient of surface tension and 2H is the curvature of the perturbed surface given by [32]:

$$2H = \frac{1}{R_o^2}\left[\xi + R_o^2 \frac{\partial^2 \xi}{\partial z^2}\right] \tag{3.6}$$

Further, the velocity components along the axis of the jet (at r = 0) must be finite. Finally, the following equation holds at the surface of the jet:

$$\frac{D\xi}{Dt} = \frac{\partial \xi}{\partial t} = V_r\Big|_{R_0+\xi} = V_r\Big|_{r\sim R_0} \tag{3.7}$$

Into the above equations we now introduce the constitutive equation of linear viscoelasticity, eq. (2.2). By taking the Laplace time transform of eqs. (3.1), (3.2) and (2.2) we get

$$\rho\alpha\hat{V}_r = -\frac{\partial\hat{p}}{\partial r} + \hat{\eta}(\alpha)\left\{\frac{\partial}{\partial r}\left[\frac{1}{r}\frac{\partial}{\partial r}(r\hat{V}_r)\right] + \frac{\partial^2\hat{V}_r}{\partial z^2}\right\} \tag{3.8a}$$

$$\rho\alpha\hat{V}_z = -\frac{\partial\hat{p}}{\partial z} + \hat{\eta}(\alpha)\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\hat{V}_z}{\partial r}\right) + \frac{\partial^2\hat{V}_z}{\partial z^2}\right] \tag{3.8b}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r\hat{V}_r) + \frac{\partial\hat{V}_z}{\partial z} = 0 \tag{3.9}$$

where α is the transform variable, and $\hat{\eta}(\alpha)$ is the complex viscosity of eq. (2.3) which represents the linear response of any viscoelastic material. The boundary conditions similarly become,

$$\left[\frac{\partial\hat{V}_z}{\partial r} + \frac{\partial\hat{V}_r}{\partial z}\right]_{r\sim R_0} = 0$$

$$\left[-\hat{p} + 2\hat{\eta}(\alpha)\frac{\partial\hat{V}_r}{\partial r}\right]_{r\sim R_0} = \frac{\sigma}{R_0^2}\left[\hat{\xi} + R_0\frac{\partial^2\hat{\xi}}{\partial z^2}\right] \tag{3.10}$$

$$\alpha\hat{\xi} = [V_r]_{r\sim R_0} \tag{3.11}$$

Henceforth, the solution is identical to Weber's corresponding solution for a Newtonian jet, with the constant Newtonian viscosity η_0 replaced by $\hat{\eta}(\alpha)$.

The continuity equation is satisfied by a stream function, $\hat{\psi}$, which is related to the velocity components by

$$\hat{V}_r = \frac{1}{r}\frac{\partial \hat{\psi}}{\partial z} \quad ; \quad \hat{V}_z = -\frac{1}{r}\frac{\partial \hat{\psi}}{\partial r} \tag{3.12}$$

Eliminating the pressure from the equations of motion by cross differentiation, and introducing the stream function as given by eq. (3.12) yields a linear differential equation for $\hat{\psi}$:

$$\alpha \Delta \hat{\psi} = \hat{\eta}(\alpha) \Delta \Delta \hat{\psi} \tag{3.13}$$

where Δ is a linear operator given by

$$\Delta \equiv \frac{\partial^2}{\partial r^2} - \frac{1}{r}\frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2} \tag{3.14}$$

Eq. (3.13) admits a solution in the form

$$\hat{\psi} = \left[\hat{\psi}_1(r) + \hat{\psi}_2\right] e^{ikz} \tag{3.15}$$

where $\hat{\psi}_1$ is the solution to

$$\alpha \hat{\psi}_1 = \hat{\eta}(\alpha)\left[\frac{d^2\hat{\psi}_1}{dr^2} - \frac{1}{r}\frac{d\hat{\psi}_1}{dr} - k^2\hat{\psi}_1\right] \tag{3.16}$$

and $\hat{\psi}_2$ is the solution to

$$\frac{d^2\hat{\psi}_2}{dr^2} - \frac{1}{r}\frac{d\hat{\psi}_2}{dr} - k^2\hat{\psi}_2 = 0 \tag{3.17}$$

The complete formal solution for $\hat{\psi}$ becomes

$$\hat{\psi} = r\left[C_1 I_1(kr) + C_2 I_1(\ell r) + C_3 K_1(kr) + C_4 K_1(\ell r)\right] e^{ikz} \tag{3.18}$$

where

$$\ell^2 = k^2 + \rho\alpha/\hat{\eta}(\alpha) \tag{3.19}$$

The quantity k may be interpreted as the wave number of a disturbance-wave, and is related to wave length, λ , by

$$\lambda = 2\pi/k$$

$I_n(\ )$ and $K_n(\ )$ are modified Bessel functions of order n; C_1, C_2, C_3 and C_4 are arbitrary constants. Application of the boundary conditions leads to an algebraic equation which relates α to k:

$$\alpha^2 + \frac{2\hat{\eta}(\alpha)k^2}{\rho\, I_0(kR_0)}\left[I_1'(kR_0) - \frac{2kl}{k^2+l^2}\cdot\frac{I_1(kR_0)}{I_1(lR_0)}\cdot I_1'(lR_0)\right]\alpha =$$

$$= \frac{\sigma k}{2\rho R_0^2}\,(1-k^2R_0^2)\cdot\frac{I_1(kR_0)}{I_0(kR_0)}\cdot\frac{(l^2-k^2)}{(l^2+k^2)} \qquad (3.20)$$

where $I_n'(\ )$ denotes differentiation of $I_n(\ )$ with respect to the argument.

Eq. (3.20) is the <u>characteristic equation</u> for the problem, and the complex number α is the <u>characteristic value</u> (or the <u>eigen value</u>) corresponding to a particular wave number k. Since the dynamic equations are linear, the perturbed motion is proportional to an exponential factor $e^{\alpha t}$ so that α may be regarded as a complex frequency. Depending on whether the real part of α is positive, zero, or negative the initial disturbances are amplified, neutrally stable, or damped, respectively. Solution of the characteristic equation yields the eigen value corresponding to a disturbance of a given wave length.

3.2 SOLUTION OF THE CHARACTERISTIC EQUATION

Eq. (3.20) is in general very complex (since the term ℓ appearing in the arguments of the Bessel function, is dependent on α) and can not be solved analytically. However, for small arguments the Bessel functions can be approximated by the leading terms of their series expansion †. We let

$$I_0(kR_0) \sim 1 \quad , \quad I_1(kR_0) \sim \frac{kR_0}{2} \quad , \quad I_1(\ell R_0) \sim \frac{\ell R_0}{2}$$

$$I_1'(kR_0) = I_1'(\ell R_0) \sim \frac{1}{2} \qquad (3.21)$$

These approximations correspond to conditions of practical interest in which the wave lengths of the disturbances are far greater than the unperturbed radius of the jet; i.e.

$$kR_0 << 1$$

For these approximations the characteristic equation becomes

$$\alpha^2 + \frac{3\hat{\eta}(\alpha)k^2}{\rho} - \frac{\sigma k^2}{2\rho R_0}(1 - k^2R^2) = 0 \qquad (3.22)$$

The values of α satisfying eq. (3.22), for a fixed value of k, obviously depend on $\hat{\eta}(\alpha)$ which itself is a function of α .

---

† The series expansion of $I_0(x)$ and $I_1(x)$ is given by

$$I_0(x) = 1 + \left(\frac{x}{2}\right)^2 + \frac{\left(\frac{x}{2}\right)^4}{2^2} + \frac{\left(\frac{x}{2}\right)^6}{2^3} + \cdots$$

$$I_1(x) = \left(\frac{x}{2}\right) + \frac{\left(\frac{x}{2}\right)^3}{2} + \frac{\left(\frac{x}{2}\right)^5}{2^2 \cdot 3} + \cdots$$

For convenience we recapitulate here the general form of $\hat{\eta}(\alpha)$,

$$\hat{\eta}(\alpha) = \frac{h}{\alpha} + \eta_\infty + \sum_{r=1}^{N} \frac{c_r}{\mu_r + \alpha} \tag{2.3}$$

With this expression in mind, we can now proceed to evaluate the properties of eq. (3.22) by resorting to a simple geometrical argument: For a fixed value of k, eq. (3.22) can be written as

$$\hat{\eta}(\alpha) = \frac{B}{\alpha} - B'\alpha \tag{3.23}$$

where

$$B = \frac{\sigma(1 - k^2 R_o^2)}{6 R_o} \quad , \quad B' = \frac{\rho}{3k^2}$$

Note that $B > 0$ since eq. (3.22) (and hence eq. (3.23) corresponds to the assumption that $kR_o \ll 1$. Consider first the case of a Newtonian fluid of constant viscosity $\hat{\eta}(\alpha) = \eta_o$. If we plot both sides of eq. (3.23) as functions of α (fig. 3.1), the resulting two curves always intersect at a point which corresponds to a real positive eigen value (see point 1 in fig. 3.1). This shows that a jet of a Newtonian fluid is unconditionally unstable relative to small disturbances. Indeed for a Newtonian fluid of viscosity η_o, eq. (3.22) is a quadratic in α having roots

$$\alpha = -\frac{3\eta_o k^2}{2\rho} \pm \sqrt{\left(\frac{3\eta_o k^2}{2\rho}\right)^2 + \frac{\sigma k^2}{2\rho R_o}(1 - k^2 R_o^2)} \tag{3.24}$$

one of which is always real and positive.

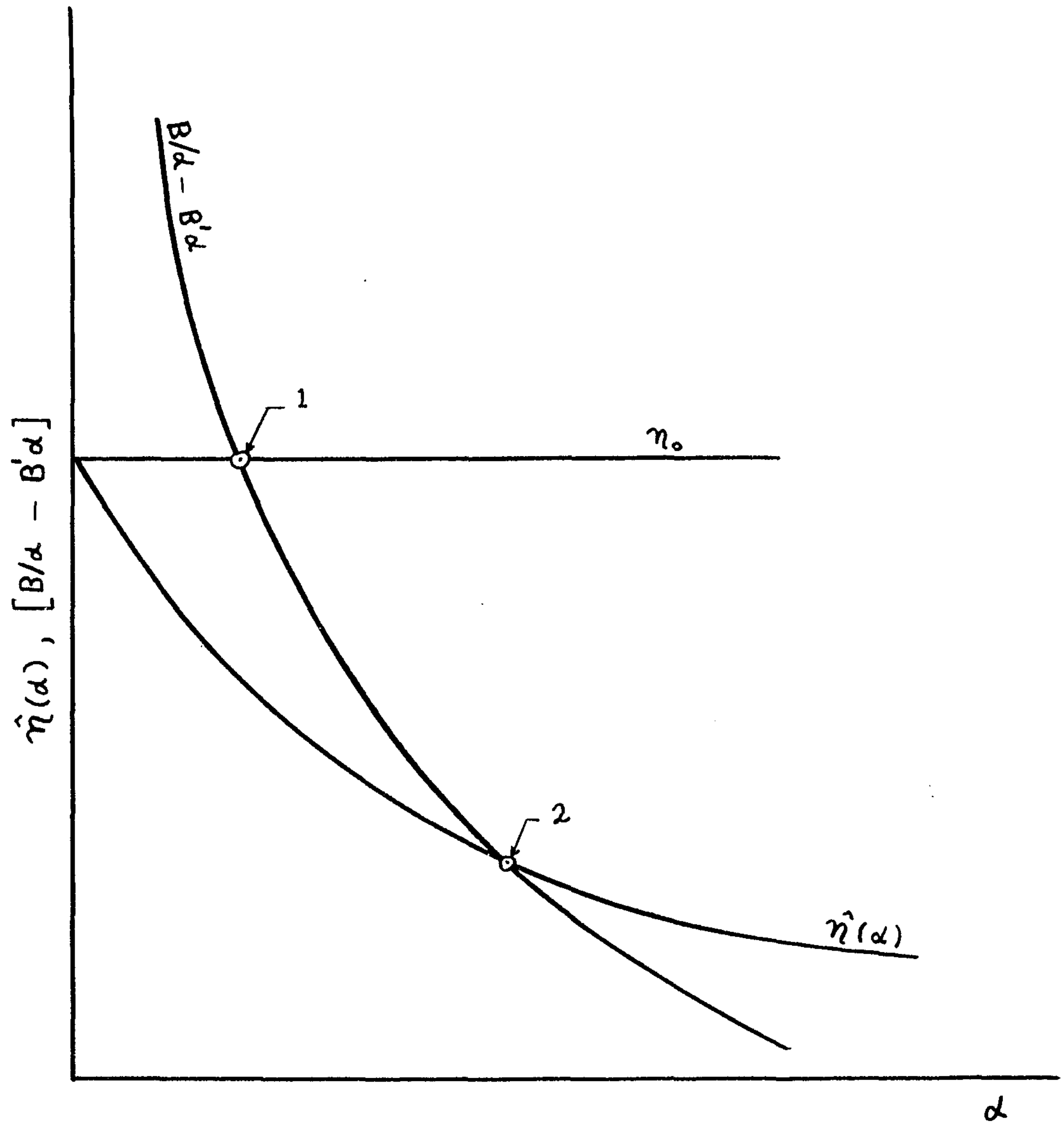

Fig. 3.1 Qualitative representation of eq. (3.23) for a Newtonian and a viscoelastic fluids.

In the case of a viscoelastic fluid (with h = 0 in eq.(2.3), $\hat{\eta}(\alpha)$ is bounded from above by

$$\hat{\eta}(0) = \eta_\infty + \sum_{r=1}^{N} C_r/\mu_r$$

and from below by η_∞. In this case a plot of both sides of eq. (3.23) as functions of α will again gives rise to an intersection between the two curves (point 2 in fig. 3.1) corresponding to a positive real eigen value. Moreover, since $\hat{\eta}(\alpha)$ decreases monotonically with α, we can conclude that, for a fixed value of k such that $kR_0 \ll 1$, the magnitude of the eigen value of a viscoelastic fluid will always <u>exceed</u> the corresponding value of a Newtonian fluid of viscosity $\eta_0 = \hat{\eta}(0)$. In fact, a plot of α vs. k^2 (derived from eq. 3.22) for a typical viscoelastic fluid with a finite $\hat{\eta}(0)$ yields a curve which lies above the curve corresponding to a Newtonian fluid of the same zero viscosity, and below the curve corresponding to an inviscid fluid ($\hat{\eta}(\alpha) \equiv 0$); with all the three curves crossing the k^2-axis at the same points (fig. 3.2). It follows that a jet of a viscoelastic fluid characterized by a complex viscosity $\hat{\eta}(\alpha)$ which is bounded by $\hat{\eta}(0) = \eta_0$ is less stable, relative to infinitesimal disturbances, than a jet of a Newtonian fluid of viscosity η_0. Any experimentally observed greater stability must be, therefore, due to non-linear phenomena or, alternatively, the fluid may not be a real fluid in the sense that it is characterized by a complex viscosity with $h \neq 0$.

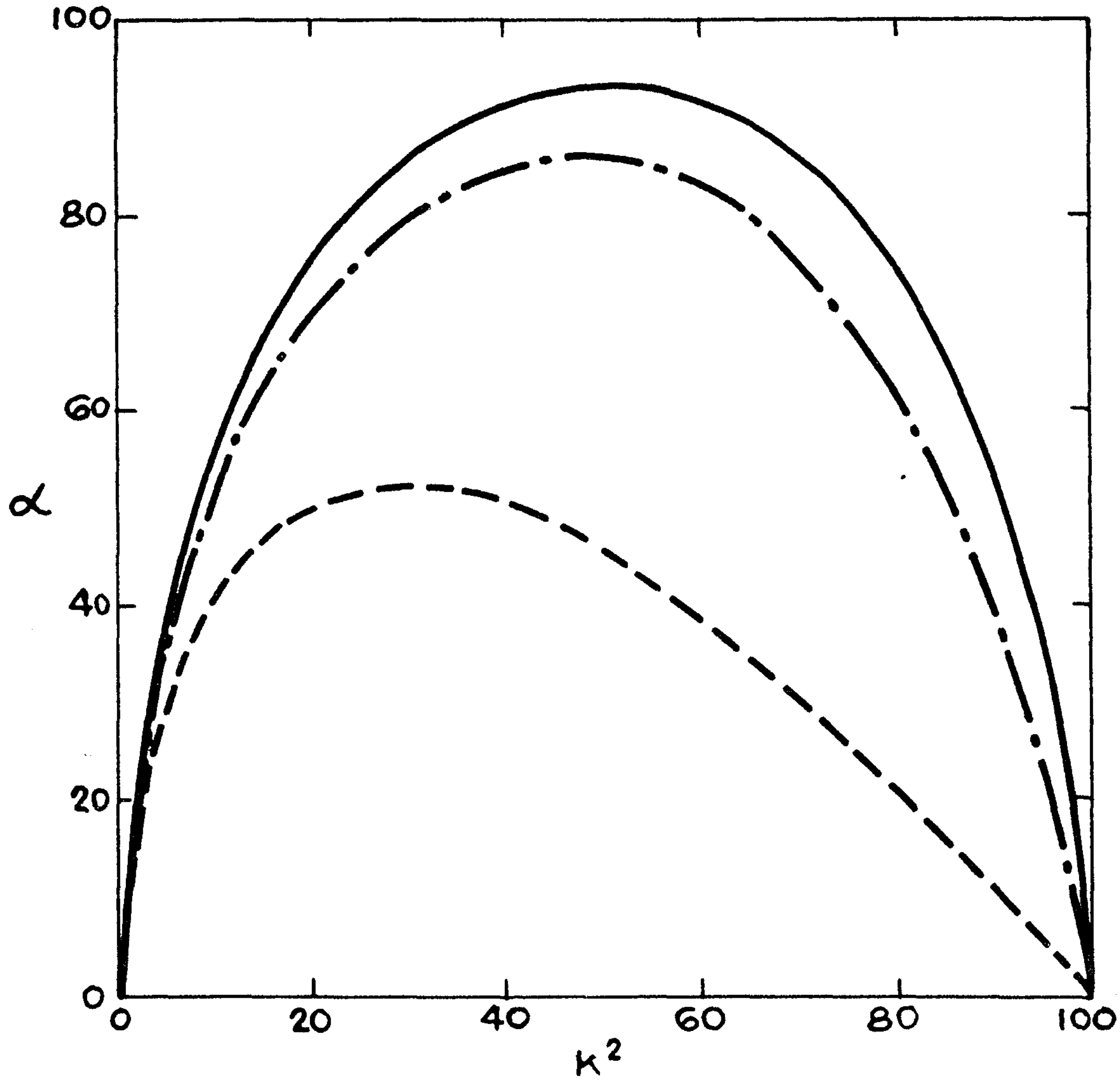

FIG. 3.2 A PLOT OF α VS. K^2 FOR:
——— INVISCID FLUID, $\hat{\eta}(\alpha) = 0$;
– – – – – NEWTONIAN FLUID,
$\eta_0 = 1$; —·—·— VISCOELASTIC FLUID, $\hat{\eta}(\alpha) = \eta_0/(1 + \lambda\alpha)$,
$\eta_0 = 1$, $\lambda = 0.1$, $\sigma = 70$, $\rho = 1$, $R_0 = 0.1$.
ALL VALUES IN CGS UNITS.

When h is finite is eq. (2.3) and all the μ_r are zero, we deal with a material which displays a purely elastic behavior, and for which

$$\hat{\eta}(\alpha) = h/\alpha \tag{3.25}$$

Referring again to eq. (3.23) and to fig. 3.3, it follows that if h < B there will always be a positive real eigen value (point 1 in fig. 3.3). When h > B, however, no positive real root exists and the jet is unconditionally stable. If $\mu_r \neq 0$ we find similarly that when h > B the jet is unconditionally stable, whereas for any viscoelastic material with h < B, there exists a positive real α causing instability. In the latter case, however, no meaningful basis of comparison can be established between the behavior of such a jet and a jet of a Newtonian fluid.

For unstable jets, droplets formation is controlled by the particular wave which grows most rapidly. The fastest growing wave can be found by implicit differentiation of eq. (3.22) and letting $d\alpha/dk = 0$ (the existence of a positive real α has already been shown). The fastest growing rate α^*, can in this way be obtained from the following expression,

$$\alpha^*\left[2 + \frac{3\hat{\eta}(\alpha^*)}{\rho}\sqrt{\frac{2\rho}{\sigma R_o}}\right] = \sqrt{\frac{\sigma}{2\rho R_o^3}} \tag{3.26}$$

The corresponding wave number is obtained from,

$$k^{*2} = \alpha^*\left[2\rho/\sigma R_o\right]^{1/2} \tag{3.27}$$

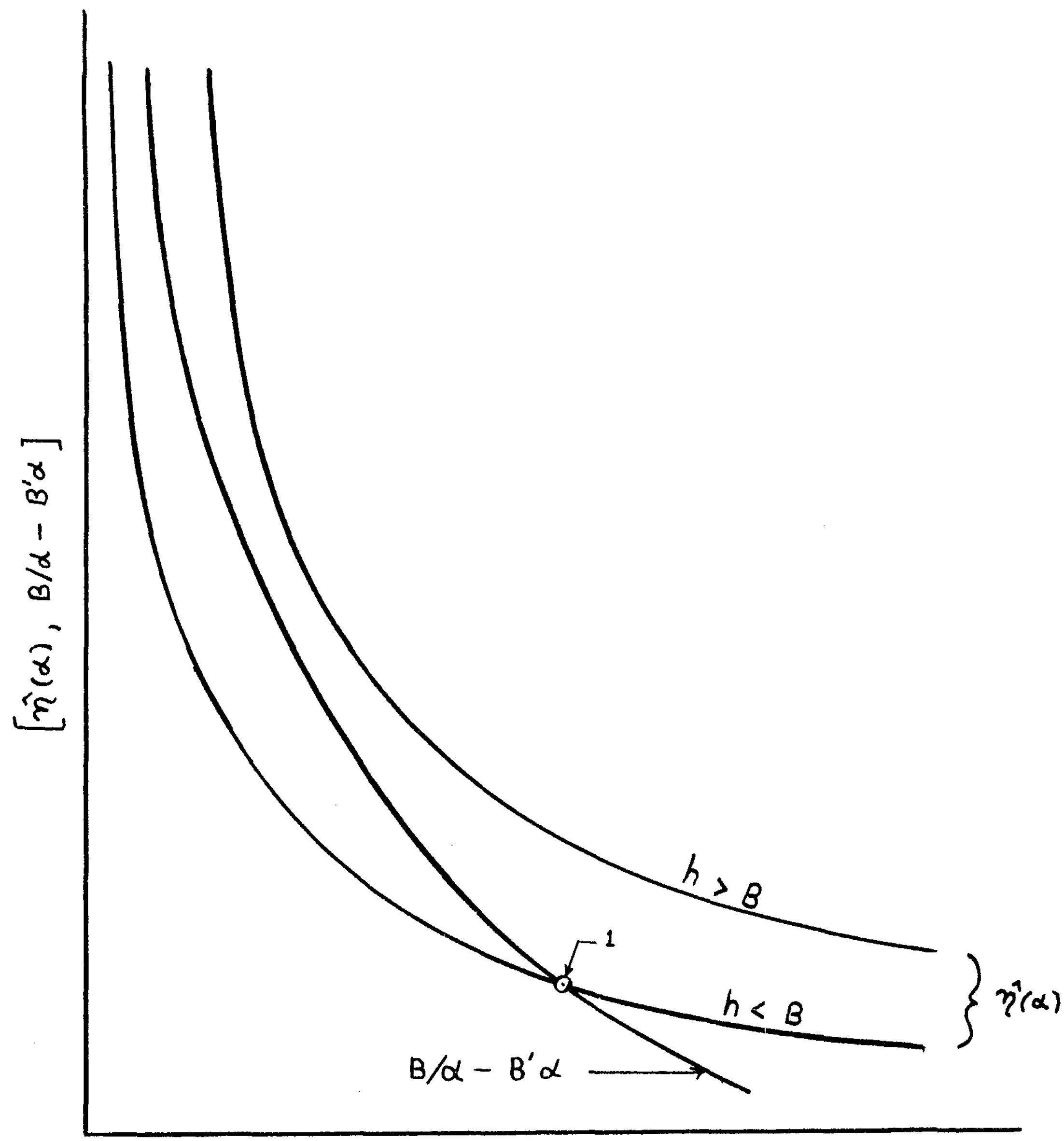

Fig. 3.3 Qualitative representation of eq. (3.23) for a purely elastic material of modulus h.

For a Newtonian fluid $\hat{\eta}(\alpha) = \eta_o$, and eqs. (3.26) and (3.27) reduce to

$$\alpha_o^* = \frac{\sqrt{\dfrac{\sigma}{2\rho R_o^3}}}{2 + \dfrac{3\eta_o}{\rho}\sqrt{\dfrac{2\rho}{\sigma R_o}}} \tag{3.28}$$

$$k_o^* = \left[3\eta_o R_o \sqrt{\frac{2R_o}{\sigma\rho}} + 2R_o^2\right]^{-1/2} \tag{3.29}$$

Eqs. (3.28) and (3.29) are identical to the expressions obtained by Weber [71]. The zero subscript in α_o^* and k_o^* denotes Newtonian values.

For Newtonian fluids of low viscosity (such as water) for which

$$2 >> \frac{3\eta_o}{\rho}\sqrt{\frac{2\rho}{\sigma R_o}} \tag{3.30}$$

the fastest growing rate is given by

$$\alpha_{in}^* = \sqrt{\frac{\sigma}{8\rho R_o^3}} \tag{3.31}$$

which is identical to the result obtained by Rayleigh for an inviscid fluid; α^*_{in} is accordingly designated by the subscript "in". The corresponding wave length is

$$k_{in}^* = 0.707/R_o \tag{3.32}$$

For Newtonian fluids of high viscosity such that

$$2 << \frac{3\eta_o}{\rho}\sqrt{\frac{2\rho}{\sigma R_o}} \tag{3.33}$$

the fastest growing rate is given by

$$\alpha_o^* = \sigma/(6 R_o \eta_o) \tag{3.34}$$

No simple expression for α^* can be obtained for a jet of a viscoelastic fluid in view of the complexity of $\hat{\eta}(\alpha)$. The value of α^* can, however,be readily computed from eq. (3.26) when $\hat{\eta}(\alpha)$ assumes a simple form. Thus, for example, for a jet of a viscoelastic fluid for which

$$\hat{\eta}(\alpha) = \frac{\eta_0}{1 + \lambda\alpha} \tag{3.35}$$

the fastest growing rate is the positive root of the following quadratic equation:

$$2\lambda\alpha^{*2} + \left[2 + \frac{3\eta_0}{\rho}\sqrt{\frac{2\rho}{\sigma R_0}} - \lambda\sqrt{\frac{\sigma}{2\rho R_0^3}}\right]\alpha^* - \sqrt{\frac{\sigma}{2\rho R_0^3}} = 0 \tag{3.36}$$

The relative stability of jets of viscoelastic and Newtonian fluids can now also be compared by means of eq. (3.26). For this purpose eq. (3.26) is divided through by

$$2 + \frac{3\eta_0}{\rho}\sqrt{\frac{2\rho}{\sigma R_0}}$$

Using eq. (3.28) then yields

$$\frac{\left[2 + \frac{3\hat{\eta}(\alpha^*)}{\rho}\sqrt{\frac{2\rho}{\sigma R_0}}\right]}{\left[2 + \frac{3\eta_0}{\rho}\sqrt{\frac{2\rho}{\sigma R_0}}\right]} = \frac{\alpha_0^*}{\alpha^*} \tag{3.37}$$

It follows from equation (3.37) that the value of the ratio α_0^*/α^* depends on the relative magnitude of $\hat{\eta}(\alpha^*)$ with respect to η_0. For a viscoelastic fluid for which $\hat{\eta}(\alpha)$ is bounded by $\hat{\eta}(0)$, $\hat{\eta}(\alpha^*) < \hat{\eta}(0)$. Consequently, it follows from eq. (3.37) that $\alpha_0^*/\alpha^* < 1$, and hence, that a jet of such a fluid is less stable than a Newtonian jet of viscosity $\eta_0 = \hat{\eta}(0)$.

3.3 DISCUSSION OF THE RESULTS

It has been shown that if the instability of a viscoelastic jet is dominated by a real latent root α^*, and if the complex viscosity is bounded in absolute value by η_0, then the jet is less stable than a jet of a Newtonian fluid of viscosity η_0. On the other hand, when $\hat{\eta}(\alpha)$ is unbounded the jet may become completely stable. The analysis which led to these conclusions is subjected to several assumptions whose validity should be assessed before the results can be evaluated in the light of the experimental evidence.

One of the underlying assumptions in the present analysis is that the jet is completely relaxed initially. In reality, the free jet experiences a stress relaxation in a region near the exit nozzle. Whereas for low-speed Newtonian jets this region is negligibly short in length, the corresponding length for viscoelastic jets may be significant and thus strongly affect the subsequent stability of these jets. The use of a constitutive equation in which $\hat{\eta}(\alpha)$ assumed the form given by eq. (2.3) is valid only when the initial profile is virtually relaxed. Should the stress relaxation occur along a significant portion of the jet, the use of this equation ceases to be valid. Furthermore, in contrast to a Newtonian fluid, the flow of a viscoelastic fluid inside the capillary tube is accompanied be generation of normal stresses; the decay of these stresses along the free jet can not be accounted for by the linear theory. In the final analysis, one has to weigh the assumption regarding the initial relaxed profile in the light of the experimental conditions and the properties of the particular fluid of interest.

The next obvious question bears upon the validity of a linear stability analysis. The present analysis presupposes the existence of wave-like disturbances whose initial amplitudes are of infinitesimal order of magnitude as compared to the unperturbed radius of the jet. The stability of the jet is, therefore, analized relative to these disturbances only. Consequently, one can neglect the non-linear terms in the equations of motion and, at the same time, employ a linear constitutive equation in place of a more general, non-linear, equation. If in the course of their growth, the amplitudes of the disturbances attain a moderate size, the linear analysis can no longer be considered applicable and the subsequent behavior of the jet must be investigated with due account to non-linear effects.

In the case of low-speed Newtonian jets, the linear analysis leads to results which are in excellent agreement with the experimental data despite the fact that the amplitude of the disturbance-wave does attain a moderate size.

This may seem to indicate that the non-linear effects which are inherent in the momentum equations do not alter significantly the growth of disturbances on the surface of low-speed Newtonian jets.

On the other hand, the break-up of a jet of a strongly viscoelastic fluid is characterized by the "droplet-thread" formation (fig. 1.2, p. 2), a phenomenon which seems to lie outside the range of the linear analysis. This does not rule out the validity of the linear analysis. The linear analysis merely attempts to describe the stability of the jet relative to infinitesimal disturbances; in this sense it is completely valid. The subsequent growth of the initial disturbances and the final shape of the jet prior to break-up are, in principle, governed by non-linear mechanism and should not necessarily reflect the results of the linear analysis. Yet, it is interesting to note that the appearance of the first, well-defined droplet during the break-up of a viscoelastic jet always occurs at a distance from the nozzle which is shorter than the break-up length of a Newtonian jet of comparable zero viscosity [27]. This may very well be a reflection of the results of the linear analysis.

Even more significant, in relation to the linear analysis, are the results of Goldin [27] for jets of weakly viscoelastic fluids. Here a sinusoidal wave formation is initially apparent.

However, the subsequent growth pattern is altered before break-up and a string of droplets connected by thin filaments of fluid is gradually formed. The droplets are spaced at relatively uniform intervals indicating that they have originated from a constant-wave-length disturbance, in accordance with the results of the linear analysis. By redefining the break-up length as the distance from the nozzle to the point where the amplitude of the disturbance is comparable to the original jet radius, Goldin observed that these jets are less stable than Newtonian jets, with the comparison based on equivalent zero viscosity. Furthermore, the relative values of the wave lengths (represented in the case of the viscoelastic jet by the distance between two adjacent droplets) are also in agreement with the results of the linear analysis.

Thus, although the instability of viscoelastic jets is largely dominated by non-linear phenomena, some relevance to the linear analysis is apparent. Yet, the linear analysis fails to account for the most striking phenomenon, namely, the formation and the unusual stability of the liquid filaments which connect the large droplets. This analysis is indeed inappropriate since the filaments appear to be under axial tension and are continually thinning. The formation of liquid filaments under tension and their stability relative to small surface disturbances will, hence, be examined in the next two chapters.

Finally, the results of the forgoing analysis indicate that a jet of a viscoelastic fluid which is characterized by a complex viscosity that is not bounded as $\alpha \longrightarrow 0$ (i.e. $h = 0$ in eq. 2.3) may be completely stable. However, since no conclusive evidence bearing upon this point is available, we are not in a position to offer any comprehensive evaluation of this result. Moreover, a reliable apparatus for measuring the complex viscosities of viscoelastic solutions in the low-frequency range has not yet been developed. Hence, most of the zero viscosity data reported on in the literature is based on extrapolated values. It is, therefore, very conceivable that some viscoelastic solutions that possess an internal structure may exhibit an instantaneous elastic response at low frequencies. According to our results, the stability of jets of these solutions would be enhanced.

CHAPTER 4

THE SPINNING OF A FILAMENT OF FLUID; MECHANICAL ASPECTS

4.1 INTRODUCTION

In the previous chapter we noted the formation of thin liquid filaments which occurs during the break-up of jets of viscoelastic fluids. In the case of Newtonian jets, filament formation is virtually absent; filaments of Newtonian fluids which have occasionally been observed are relatively short in length and break up rapidly into secondary droplets. This contrasting behavior is undoubtedly a manifestation of the differences in the rheological properties of viscoelastic and Newtonian fluids.

It is our purpose in this chapter to examine the effect of the rheological properties of various fluids on their ability to form liquid filaments. A direct analysis of the formation of liquid filaments during the break-up of capillary jets is not possible since we do not possess, at present, sufficient information which would enable us to simulate the conditions under which these filaments are formed. Instead, the analysis will be confined to a comparable phenomenon, namely, the formation of filaments in spinning processes. In both cases, the steady behavior of a liquid filament under axial tension represents the most essential feature. The spinning process, admittedly, involves other complex meachnisms; however, the conclusions that can be drawn from the analysis of the mechanical aspects of the spinning process can be construed with respect to the formation of liquid filaments in atomization processes and other related applications.

In a typical commercial process for the production of synthetic fibers (fig. 4.1), the fluid is forced under pressure through a symmetrically arranged set of holes in a flat plate, the spinnerette. The resulting threads are then rapidly pulled away by a wind-up roll situated as some distance L from the spinnerette.

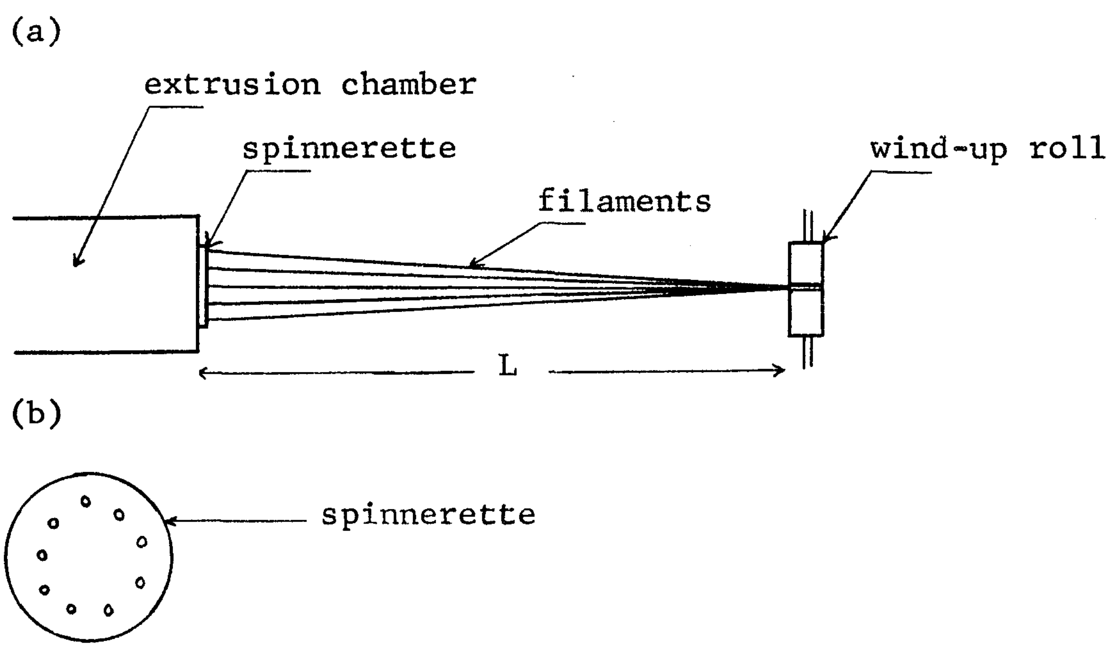

Fig. 4.1 Schematic diagram of the spinning process:

(a) General view; (b) the spinnerette.

Several spinning methods are currently used, all of which are designed to produce self-supporting filaments of sufficient strength and hardness. The fluid is generally fed into the extrusion chamber under conditions which facilitate the extrusion of the filaments through the narrow holes of the spinnerette (of the order of 10^{-2}cm in diameter). The transition of the fluid from conditions which prevail in the extrusion chamber to self-supporting filaments is accompanied by a great increase in the viscosity of the fluid. This hardening phenomenon is accomplished by a suitable choice of the medium (and its conditions) through which the filaments are drawn. Accordingly, the spinning methods can be classified into three broad categories:

(1) Melt Spinning

The filament material, generally a polymer, is extruded in a melt form into an atmosphere (air) having a temperature lower than the melting temperature range of the polymer. Typical drawing speeds are of the order of 10^3cm/sec .

(2) Dry Spinning

The fluid is dissolved in a suitable volatile solvent prior to its introduction into the extrusion chamber. The filaments are drawn through air with hardening being caused by solvent evaporation. Typical drawing speeds are of the order of 10^3 cm/sec .

(3) Wet Spinning

The filament fluid in a suitable solution is extruded into a bath of liquid of consistency similar to that of water. Hardening is caused by solvent diffusion into the surrounding fluid. In contrast to Dry and Melt Spinning, the drawing speeds in a Wet Spinning process are reduced to an order of 10^2 cm/sec due to the drag which the surrounding liquid exerts upon the filament.

The term spinnability (or "Spinnbarkeit") has often been used in the literature to describe the ability of a fluid to form filaments. This, however, is a rather loose definition. The formation of self-supporting fibers depends both on the rheological properties of the filament fluid as well as on the choice of the proper spinning conditions. As was pointed out by Roberts [56], an "unspinnable fluid" might become "spinnable" by a modification of the spinning conditions. It is perhaps more meaningful, therefore, to define spinnability on the sole basis of the rheological properties of the fluid. In this sense a fluid may be regarded as spinnable if it can form filaments without hardening. This is not to say that such a fluid may be processed commercially without hardening. Indeed, on a commercial basis, hardening is necessary for the fulfillment of certain physical and textile requirements among which, of course, is a suitable tensile strength. However, it is very well known that certain fluids are inherently more suitable to the spinning process while others are completely inadequate regardless of the spinning method. Such a basic difference in the adaptability of various fluids to the spinning process must, ultimately, be related to their rheological properties. Unfortunatley, the enormous increase in the viscosity of the filament due to the hardening mechanism dominates the spinning process and tends to overshadow the effect of the inherent rheological properties of the fluid upon the formation of fibers derived from it. A rigorous mathematical description of the spinning process requires the simultaneous application of energy, mass and momentum balances. The resulting set of equations can not be solved at present. Therefore, the purpose of this chapter is to examine the mechanical aspects of the spinning process only. In particular, we wish to ascertain the effect of the rheological properties of the filament fluid upon its spinnability.

Considered even alone, the mechanical aspects of the spinning process represent a formidable mathematical task. In addition to the non-linearity of the dynamic equations, the main difficulty is associated with the free boundary of the filament. The free streamlines are not known in advance and must be determined as part of the solution. Further, the boundary conditions at the free surface are themselves dependent on the unknown orientation of the free streamlines. The problem can be simplified significantly by recognizing that, except for a relatively short region near the spinnerette hole, the transport of momentum at any section of the filament occurs predominantly in the axial direction. That is, the mechanics of the flow are governed largely by the balance between the axial inertia caused by the action of the wind-up roll, the resultant tensile force and the external drag. Under these conditions, the axial velocity can be considered independent of the radial direction and the momentum equations can then be reduced to an ordinary differential equation which relates the axial velocity to the spinning distance.

In the following sections, the mechanical spinning problem is first formulated in its complete form. The reduction of the equations is then accomplished with the aid of the simplifying assumptions discussed above. Solutions to the reduced problem are subsequently obtained for Newtonian and non-Newtonian fluids.

4.2 FORMULATION OF THE SPINNING PROBLEM

Assuming the flow to be steady and axisymmetric, the equations of motion and continuity are:

r-direction

$$\rho\left(v_r \frac{\partial v_r}{\partial r} + v_z \frac{\partial v_r}{\partial z}\right) = -\frac{\partial p}{\partial r} + \frac{1}{r}\frac{\partial}{\partial r}(\tau_{rr} r) + \frac{\partial \tau_{rz}}{\partial z} - \frac{\tau_{\theta\theta}}{r} \tag{4.1}$$

z-direction

$$\rho\left(v_z \frac{\partial v_z}{\partial z} + v_r \frac{\partial v_z}{\partial r}\right) = -\frac{\partial p}{\partial z} + \frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rz}) + \frac{\partial \tau_{zz}}{\partial z} + \rho g \tag{4.2}$$

continuity

$$\frac{1}{r}\frac{\partial}{\partial r}(r v_r) + \frac{\partial v_z}{\partial z} = 0 \tag{4.3}$$

The boundary conditions specify the value of the axial velocity at both ends of the spinning way and the stress distribution on the free surface of the filament. The former conditions are simply,

$$v_z(z=0,r) = V_o \tag{4.4a}$$

$$v_z(z=L,r) = V_L \tag{4.4b}$$

Turning now to the free surface, let R(z) be the radius of the filament, and $\underset{\sim}{n}$ and $\underset{\sim}{t}$ the normal and tangential unit vectors, respectively, at a point on the surface (fig. 4.2). From elementary geometry it follows that

$$\underset{\sim}{n} = \frac{\underset{\sim}{i}_r}{q} - \frac{\underset{\sim}{i}_z}{q} R' \tag{4.5a}$$

and

$$\underset{\sim}{t} = -\frac{\underset{\sim}{i}_r R'}{q} - \frac{\underset{\sim}{i}_z}{q} \qquad (4.5b)$$

where $R' = dR/dz$, $q = \left[1 + (R')^2\right]^{1/2}$ and $\underset{\sim}{i}_r$ and $\underset{\sim}{i}_z$ are the orthonormal base vectors in the radial and axial directions respectively.

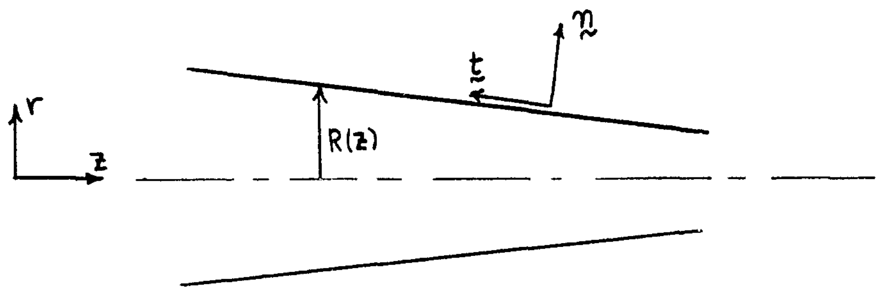

Fig. 4.2 Orientation of the free surface of the filament.

The dynamic conditions prevailing at the free surface are determined by the interaction between the filament and its surrounding. Accordingly, the normal component of the stress vector is balanced against the stress induced by the surface tension force, and the tangential component of the stress vector is balanced by the external drag caused by the surrounding medium. Denoting the magnitudes of the normal and the tangential components of the stress vector by s_{nn} and s_{nt} respectively, the above conditions are

$$s_{nn} = \underset{\sim}{n}\cdot(\underset{\sim}{n}\cdot S) = \frac{1}{q^2}\left[-p + T_{rr} - R'T_{rz}\right] + \frac{R'}{q^2}\left[T_{rz} - R'(-p + T_{zz})\right] = $$
$$= 2H\sigma \qquad (4.7)$$

and

$$s_{nt} = \underset{\sim}{t}\cdot(\underset{\sim}{n}\cdot\underset{\sim}{S}) =$$

$$= \frac{-R'}{q^2}\left[-p + T_{rr} - R'T_{rz}\right] - \frac{1}{q^2}\left[T_{rz} - R'(-p + T_{zz})\right] = F_d \qquad (4.8)$$

where σ is the coefficient of surface tension, and 2H is the mean curvature of the free surface, given by

$$2H = \frac{-1}{qR} + \frac{R''}{q^3} \qquad (4.9)$$

F_d is the external drag. A suitable expression for F_d will be incorporated into the reduced problem which is considered in the following section. At present we leave it unspecified. Multiplying Eq. (4.7) by R' and adding the resulting expression to eq. (4.8) gives

$$T_{rz} - R'(-p + T_{zz}) = -F_d - 2H\sigma R' \qquad (4.10)$$

Multiplying eq. (4.8) by -R' and adding it to eq. (4.7) gives

$$-p + T_{rr} - R'T_{rz} = -F_d R' + 2H\sigma \qquad (4.11)$$

Eqs. (4.10) and (4.11) will, henceforth, be regarded as the boundary conditions at the free surface. For convenience, we summarize the four boundary conditions for the spinning flow:

(1) $$v_z(z=0,r) = V_o \qquad (4.12a)$$

(2) $$v_z(z=L,r) = V_L \qquad (4.12b)$$

(3) $$\left[T_{rz} - R'(-p + T_{zz})\right]_{r=R} = -F_d - 2H\sigma R' \qquad (4.12c)$$

(4) $$\left[-p + T_{rr} - R'T_{rz}\right]_{r=R} = -F_d R' + 2H\sigma \qquad (4.12d)$$

In addition the following relation holds at the free surface of the filament

$$v_r(r=R) = \frac{DR}{Dt} = v_z(r=R)R' \qquad (4.13)$$

4.3 THE REDUCED PROBLEM

The following conditions are assumed:

$$\text{(i)}\ v_z >> v_r$$

$$(4.14)$$

$$\text{(ii)}\quad \frac{\partial v_z}{\partial r} = 0$$

Using condition (i), it follows from eq. (4.13) that

$$R' << 1 \qquad (4.15)$$

Using condition (ii), it follows from the continuity equation (4.3) that

$$v_r = -\frac{r}{2}\frac{\partial v_z}{\partial z} \qquad (4.16)$$

and, further, that

$$\rho \int_0^{2\pi}\int_0^R v_z\, r\, dr\, d\theta = \rho\pi v_z R^2 = \text{CONSTANT} = W \qquad (4.17)$$

Eq. (4.17) thus becomes the continuity equation for the reduced problem with W being the mass flow rate across any section of the filament. The value of W can be obtained from

$$W = \rho \pi V_o R_o^2 \tag{4.18}$$

where R_o is the radius of the spinnerette hole and V_o, the average exit velocity.

The reduction of the momentum equations (in accordance with the above simplifications) can be facilitated by an order of magnitude estimate. However, in order to assign a magnitude to the stress terms appearing in the momentum equations, it is first necessary to relate these stresses to the motion. Accordingly, the magnitude of the stress terms will be estimated on the basis of the constitutive equations for a Newtonian fluid. That is, with δ and L corresponding, respectively, to some characteristic lengths in the radial and axial directions, let

$$\begin{aligned}
T_{zz} &= 2\eta_o \frac{\partial v_z}{\partial z} = O[\eta_o V_o/L] \\
T_{rr} &= 2\eta_o \frac{\partial v_r}{\partial r} = -\eta_o \frac{\partial v_z}{\partial z} = O[\eta_o V_o/L] \\
T_{rz} &= \eta_o \frac{\partial v_r}{\partial z} = -\frac{\eta_o}{2} r \frac{\partial^2 v_z}{\partial z^2} = O\left[\frac{\delta}{L} \eta_o V_o/L\right] \\
T_{\theta\theta} &= 2\eta_o \frac{v_r}{r} = -\eta_o \frac{\partial v_z}{\partial z} = O[\eta_o V_o/L]
\end{aligned} \tag{4.19}$$

In the above equations use has been made of eq. (4.16) and the fact that $v_z = O(V_o)$. Using eq. (4.19), the z-direction momentum equation (4.2) becomes (with the magnitude of the terms which involve velocity component written below),

$$\rho V_z \frac{\partial V_z}{\partial z} = -\frac{\partial p}{\partial z} + \eta_0 \frac{\partial^2 V_z}{\partial z^2} + \rho g \tag{4.20}$$

$$O[\rho V_0^2/L] \qquad\qquad O[\eta_0 V_0/L^2]$$

Likewise, the r-direction momentum equation (4.1) becomes,

$$\rho\left[\frac{r}{4}\left(\frac{\partial V_z}{\partial z}\right)^2 - \frac{r}{2} V_z \frac{\partial^2 V_z}{\partial z^2}\right] = -\frac{\partial p}{\partial r} - \frac{r}{2}\eta_0 \frac{\partial^3 V_z}{\partial z^3} \tag{4.21}$$

$$O\left[\frac{\delta}{L}\rho V_0^2/L\right] \qquad\qquad O\left[\frac{\delta}{L}\rho V_0/L^2\right]$$

In eq. (4.21) each term which involves a velocity component is, by comparison to the corresponding term of eq. (4.20), of lesser order by a factor of δ/L. Since our basic assumptions imply that $\delta/L \ll 1$ (eq. 4.15), it follows that $\partial p/\partial r$ may be neglected in comparison to $\partial p/\partial z$. That is, the pressure is constant throughout the thickness of the filament and is dependent on the axial direction only.

The momentum equations now read:

$$\rho V_z \frac{\partial V_z}{\partial z} = -\frac{\partial p}{\partial z} + \frac{\partial T_{zz}}{\partial z} + \frac{1}{r}\frac{\partial}{\partial r}(r T_{rz}) + \rho g \tag{4.22}$$

$$0 = -\frac{\partial p}{\partial r} \tag{4.23}$$

The boundary conditions at the free surface are similarly reduced to

$$\begin{aligned} &(3) \qquad \left[T_{rz} - R'(-p + T_{zz})\right]_{r=R} = - F_d + \frac{R'\sigma}{R} \\ &(4) \qquad \left[-p + T_{rr}\right]_{r=R} = - \frac{\sigma}{R} \end{aligned} \qquad (4.24)$$

In order to take account of these conditions (since the r-direction momentum equation has been eliminated), eq. (4.22) is integrated over the cross sectional area of the filament as follows

$$\int_0^R \rho v_z \frac{\partial v_z}{\partial z} r\,dr = \int_0^R \frac{\partial}{\partial z}(-p + T_{zz}) r\,dr + \int_0^R \frac{1}{r}\frac{\partial}{\partial r}(r T_{rr}) r\,dr + \int_0^R \rho g\, r\,dr \qquad (4.25)$$

or

$$\int_0^R \rho v_z \frac{\partial v_z}{\partial r} r\,dr = \frac{d}{dz}\int_0^R (-p + T_{zz}) r\,dr + R\left[T_{rz} - (-p + T_{zz})R'\right]_{r=R} + \rho g \frac{R^2}{2} \qquad (4.26)$$

Since v_z is regarded as a function of z only, ordinary derivatives will, henceforth, replace partial derivatives whenever the quantities involved are solely related to v_z or to its higher-order derivatives. Eq. (4.26) thus becomes

$$\rho v_z \frac{d v_z}{dz} \frac{R^2}{2} = \frac{d}{dz}\left[(-p + T_{zz})\frac{R^2}{2}\right] + R\left[T_{rz} - (-p + T_{zz})R'\right]_{r=R} + \rho g \frac{R^2}{2} \qquad (4.27)$$

By using the boundary conditions at the free surface (4.24) as well as the continuity equation for the reduced problem (4.17), eq. (4.27) assumes the form:

$$W\frac{dV_z}{dz} = \pi\frac{d}{dz}\left[\left(T_{zz} - T_{rr} - \frac{\sigma}{R}\right)R^2\right] - 2\pi R\left(F_d - \frac{R'}{R}\sigma\right) + \pi\rho g R^2 \qquad (4.28)$$

A suitable expression for the external drag, F_d, can be fashioned after the results of Cooke [17] who has obtained a formula for the drag caused by the flow of fluids along thin cylinders of arbitrary cross-section. Accordingly,

$$F_d = \frac{1}{R}\mu_m V_z\left(0.696 + 1.328\, d^{-1}\right) \qquad (4.29)$$

where

$$d = \frac{4}{R}\sqrt{\frac{\mu_m L}{\rho_m V_z}}$$

ρ_m and μ_m are the density and viscosity, respectively, of the medium liquid, and L is the length of the filament. Substituting eq. (4.29) into eq. (4.28) and using the continuity equation to eliminate terms involving R and R' gives the final form of the governing equation of motion for the reduced problem:

$$W\frac{dV_z}{dz} = \frac{d}{dz}\left[\frac{1}{V_z}\left(T_{zz} - T_{rr} - \frac{\sigma}{b}V_z^{1/2}\right)\right] - 2\pi V_z\left[0.696\mu_m + 0.332\, b\sqrt{\frac{\rho_m\mu_m}{L}}\right]$$

$$- \frac{2\pi b\sigma}{V_z^{3/2}}\frac{dV_z}{dz} + \frac{Wg}{V_z} \qquad (4.30)$$

where

$$b = \sqrt{\frac{W}{\rho\pi}}$$

The boundary conditions are:

$$V_z(0) = V_0 \quad , \quad V_z(L) = V_L \quad ; \quad V_L > V_0 \tag{4.31}$$

It should be noted that the reduction of the dynamic equations leading to eq. (4.30) has been, in part, based upon the constitutive equation for a Newtonian fluid. Eq. (4.30), nonetheless, will be regarded as the governing momentum equation for filaments of non-Newtonian fluids as well; that is, provided that the constitutive equations for such fluids do not offer any substantial contradiction to the arguments presented above.

4.4 SOLUTIONS FOR A FILAMENT OF A NEWTONIAN FLUID

For a Newtonian fluid,

$$T_{zz} = 2\eta_0 \frac{dV_z}{dz} \; , \; T_{rr} = -\eta_0 \frac{dV_z}{dz}$$

The governing equation of motion for a filament of a Newtonian fluid thus becomes

$$W\frac{dV_z}{dz} = \frac{d}{dz}\left[\frac{1}{V_z}\left(3\eta_0 \frac{dV_z}{dz} - \frac{\sigma}{b} V_z^{1/2}\right)\right] - 2\pi f V_z - \frac{2\pi b\sigma}{V_z^{3/2}}\frac{dV_z}{dz} + \frac{Wg}{V_z} \tag{4.32}$$

where

$$b = \sqrt{\frac{W}{\rho\pi}} \quad , \quad f = 0.696\mu_m + 0.332\, b\sqrt{\frac{\rho_m \mu_m}{L}}$$

The boundary conditions are,

$$V_z(0) = V_0 \quad , \quad V_z(L) = V_L \tag{4.33}$$

We now introduce the following dimensionless quantities:

$$v = \frac{V_z}{V_o} \quad ; \quad \delta = \frac{R}{R_o} \quad ; \quad \xi = \frac{z}{L} \tag{4.34}$$

In a dimensionless form, the continuity and the equation of motion become, respectively,

$$\delta^2 v = 1 \tag{4.35}$$

$$\frac{dv}{d\xi} = \frac{d}{d\xi}\left[\frac{1}{v}\left(\frac{1}{R_L}\frac{dv}{d\xi} - \Sigma v^{1/2}\right)\right] - 2Fv - \Sigma v^{-3/2} + \frac{G}{v} \tag{4.36}$$

where

$$R_L = \frac{\rho V_o L}{3\eta_o} \quad ; \quad \Sigma = \frac{1}{\rho V_o^2}\frac{\sigma}{R_o} \quad ; \quad G = \frac{gL}{R_o^2}$$

$$F = \frac{\rho_m}{\rho}\left[0.332\,\frac{L}{R_o}(R_m)^{-1/2} + 0.696\,\frac{L^2}{R_o^2}(R_m)^{-1}\right]$$

and

$$R_m = \rho_m V_o L/\mu_m$$

The boundary conditions are similarly defined as

$$v(0) = 1 \quad ; \quad v(1) = \frac{V_L}{V_o} = v_1 \quad ; \quad v_1 > 1 \tag{4.37}$$

The dimensionless groups defined above represent ratios of magnitudes between the inertia and the other forces which arise in the spinning process. Thus,

$$R_L = \frac{\text{viscous force}}{\text{inertia}}$$

$$F = \frac{\text{external drag}}{\text{inertia}}$$

$$\Sigma = \frac{\text{surface tension}}{\text{inertia}}$$

$$G = \frac{\text{gravity}}{\text{inertia}}$$

Eq. (4.36) can not be solved analytically; consequently, numerical solutions were obtained with the aid of a digital computer. However, it is worthwhile to examine in detail two special cases for which an analytic solution can be obtained.

1. Solution in the Absence of Drag, Gravity and Surface Tension

In the absence of gravity and surface effects, eq. (4.36) reduces to

$$R_L \frac{dv}{d\xi} = \frac{d}{d\xi}\left[\frac{1}{v}\frac{dv}{d\xi}\right] \tag{4.38}$$

with boundary conditions,

$$v(0) = 1 \quad ; \quad v(1) = v_1 \quad ; \quad v_1 > 1 \tag{4.39}$$

The first integral of eq. (4.38) is

$$\frac{dv}{d\xi} = R_L v(v - C) \tag{4.40}$$

where C is an arbitrary constant. Integrating eq. (4.40) between $\xi = 0$ and $\xi = \xi$ yields

$$v(\xi) = \frac{C}{(C-1)e^{R_L C\xi} + 1} \tag{4.41}$$

The value of the constant C is to be determined from the boundary condition at $\xi = 1$. It is clear from eqs. (4.40) and (4.41) that the value of C should be less than unity; otherwise, the velocity will increase along the spinning way, that is, no solution would exist. When C = 1, $v(\xi) = 1$ identically.

A close examination of eq. (4.41) shows that the boundary condition at $\xi = 1$ (i.e. $v = v_1$; $v_1 > 1$) can be satisfied for any independent choice of v_1 and R_L. In particular, we can choose R_L as large as we please (corresponding to a low-viscosity fluid), and yet obtain any value of v_1 ($v_1 > 1$) we desire. Physically this is impossible since a very low-viscosity fluid (such as water) can not be spun at all. To see that such is the case, let

$$1 - C = e^{-\gamma} \; ; \quad \gamma > 0 \tag{4.42a}$$

Eq. (4.41) becomes at $\xi = 1$,

$$v_1 = \frac{1 - e^{-\gamma}}{1 - e^{(R_L - \gamma)}} \tag{4.42b}$$

For large γ ; i.e., for a value of C very close to unity,

$$v_1 \cong \frac{1}{1 - e^{(R_L - \gamma)}} \tag{4.42c}$$

from which

$$R_L - \gamma = \ln\left(\frac{v_1 - 1}{v_1}\right) \tag{4.42d}$$

Thus, by choosing C close enough to unity, we can find a solution v_1 ($v_1 > 1$) for any large value of R_L. The choice of such a value of C is obvious from eqs. (4.42a) and (4.42d). This is a rather puzzling result. However, if we examine the variation of $v(\xi)$ along the spinning way for a large R_L (fig. 4.3), and compare it to the corresponding solution for a high-viscosity fluid (small R_L), the puzzle may be resolved. It is clear from fig. 4.3 that, for a large value of R_L, the velocity is virtually unchanged along most of the spinning way; just before $\xi = 1$, the velocity suddenly increases with the corresponding velocity gradient attaining enormously high values. This is in contrast to the case in which R_L is small, where the velocity varies gradually. The shape of the filaments corresponding to these solutions can be obtained via the continuity equation, and are depicted in fig. 4.4. Thus, for large values of R_L, the effect of pulling the filament at a given speed at $\xi = 1$ appears to be manifested only in an extremely short region near the wind-up roll; throughout most of the spinning way no appreciable elongation occurs. For such a case, however, the reduced solution is not valid since our basic assumption requires that the variation in the radius of the filament be small. One can perhaps interpret these results as an indication that filaments of low viscosity can not, indeed, be formed, since any application of

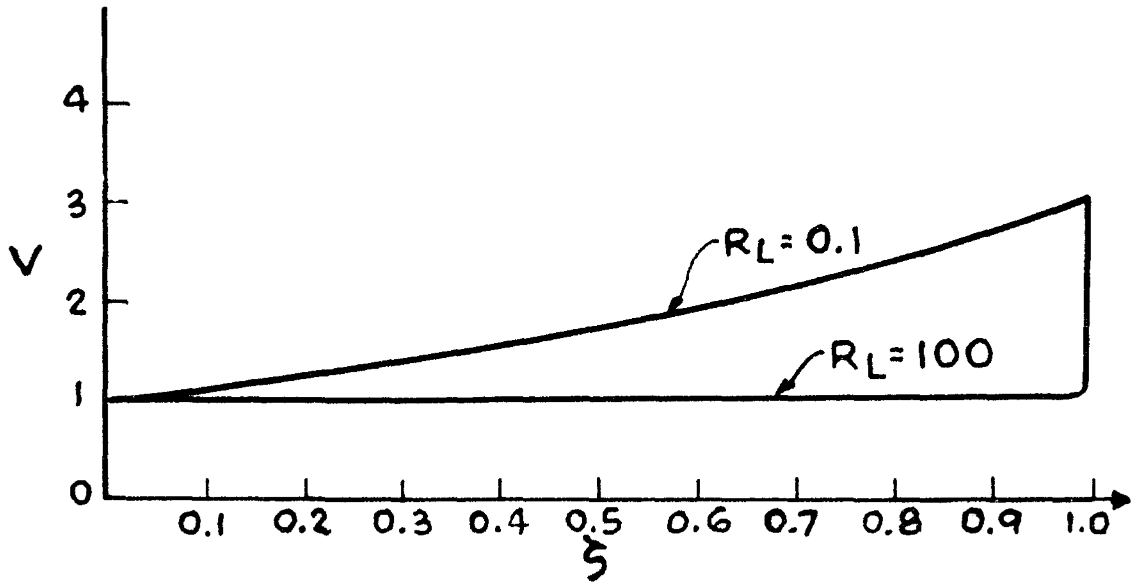

FIG. 4.3 VARIATION OF THE VELOCITY ALONG THE SPINNING WAY IN THE ABSENCE OF GRAVITY AND SURFACE EFFECTS.

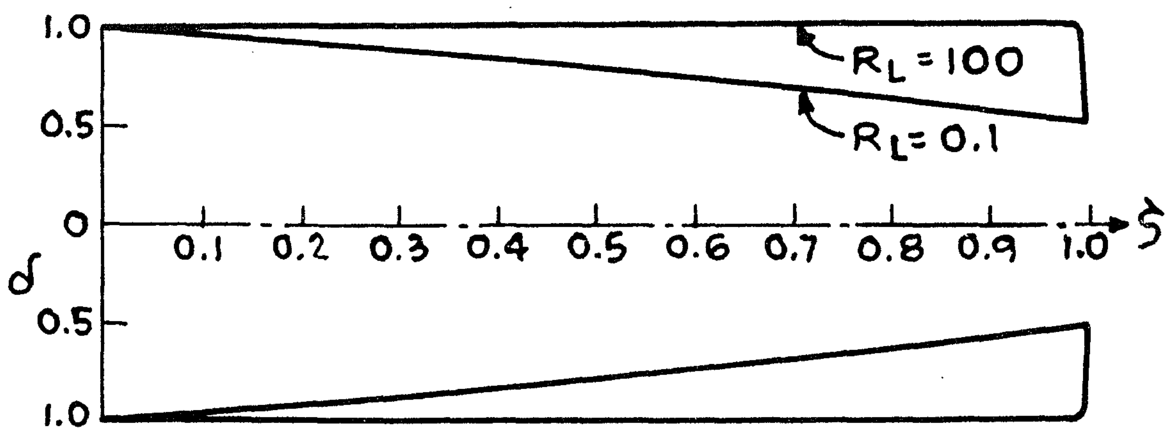

FIG. 4.4 FILAMENTS SHAPES (δ VS. ζ) CORRESPONDING TO THE VELOCITY VARIATIONS SHOWN IN FIG. 4.3.

tensile stress would result in a rapid thinning, leading to an immediate break-up of the filament.

Even in the case of high-viscosity fluids, the omission of the surface effects, especially the external drag, yields solutions which seem to furnish little insight into the spinning problem. Hence we turn now to the case in which the effect of the external drag is included.

2. <u>The Effect of the External Drag; Wet Spinning</u>.

In the interest of obtaining an analytic solution, we consider the case in which the inertial term can be neglected in comparison with external drag; i.e., for $F \gg 1$. Such a choice, in fact, represents typical conditions which prevail in most Wet Spinning processes where the filaments are drawn at a relatively low speed (of the order of 10^2 cm/sec) through a bath of liquid of consistency similar to that of water. For this case, the governing equation becomes (with gravity and surface tension not included):

$$\frac{dv}{d\xi}\left(\frac{1}{v}\frac{dv}{d\xi}\right) = 2FR_L v \tag{4.43}$$

with boundary conditions,

$$v(0) = 1 \; ; \quad v(1) = v_1 \; ; \quad v_1 > 1 \tag{4.44}$$

The solution to eq. (4.43) is

$$v(\xi) = A^2\left[1 + \tan^2\left(\frac{A\beta}{2}\xi + \tan^{-1}\sqrt{\frac{1-A^2}{A^2}}\right)\right] \tag{4.45}$$

where $\beta = 2(FR_L)^{\frac{1}{2}}$, and A is an arbitrary constant to be determined from the boundary condition at $\xi = 1$, i.e.,

$$v_1 = A^2\left[1 + \tan^2\left(\frac{A\beta}{2} + \tan^{-1}\sqrt{\frac{1-A^2}{A^2}}\right)\right] \tag{4.46}$$

For a physically meaningful solution to exist, the value of A can not exceed unity. When A= 1, eq. (4.45) becomes

$$v(\xi) = 1 + \tan^2(\beta/2)\xi \tag{4.47}$$

and v_1 is

$$v_1 = 1 + \tan^2\frac{\beta}{2} \tag{4.48}$$

For a fixed value of β, the pullaway speed, v_1, of eq. (4.48) is smaller than any v_1 of eq. (4.46) corresponding to a value of A smaller than unity, i.e.,

$$\left(1+\tan^2\frac{\beta}{2}\right) < A^2\left[1 + \tan^2\left(\frac{A\beta}{2} + \tan^2\sqrt{\frac{1-A^2}{A^2}}\right)\right] \quad ; \; A<1 \tag{4.49}$$

Similarly, $v(\xi)$ of eq. (4.47) is always smaller than the corresponding $v(\xi)$ of eq. (4.45) with $A < 1$. This is shown schematically in fig. 4.5.

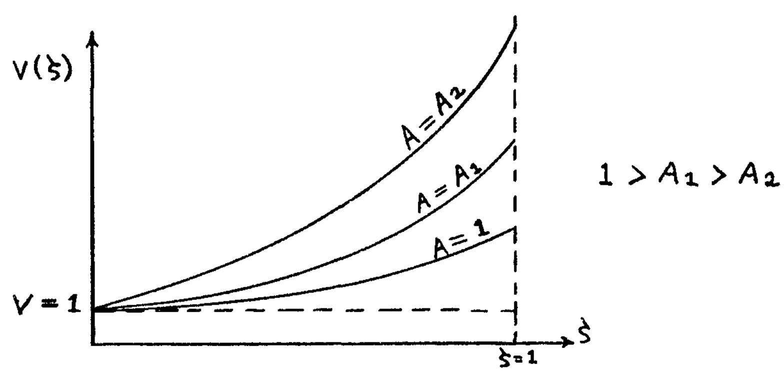

Fig. 4.5 Schematic illustration of the solutions to eq. (4.45) for different pullaway velocities.

It can be noted further that A = 1, (eq. 4.47), corresponds to the condition $dv/d\xi = 0$ at $\xi = 0$, while $A < 1$ in eq. (4.45) implies that $dv(0)/d\xi > 0$. Thus, the velocity curve of eq.(4.47) can be regarded as a bound on all existing solutions. In particular, for a fixed value of β , a solution exists only if

$$\beta < \pi \tag{4.50}$$

When $\beta > \pi$, $v(\xi)$ is singular at $\xi < 1$ and, hence, no solution exists. In terms of the flow parameters, the inequality of (4.50) is

$$\frac{\rho V_o L}{3\eta_o} \cdot \frac{\rho_m}{\rho} \left[0.332 \frac{L}{R_o} R_m^{-1/2} + 0.696 \frac{L^2}{R_o^2} R_m^{-1} \right] < \frac{\pi^2}{4} \tag{4.51}$$

where

$$R_m = \frac{\rho_m V_o L}{\mu_m}$$

Eq. (4.51) can provide an estimate of the minimum viscosity, $(\eta_o)_{min}$, which is required for some specific spinning conditions (R_o, L, etc'). Accordingly,

$$(\eta_o)_{min} = \frac{4\rho V_o}{3\pi^2} \left(\frac{\rho_m}{\rho}\right) \left[0.332 \frac{L}{R_o} R_m^{-1/2} + 0.696 \frac{L^2}{R_o^2} R_m^{-1} \right] \tag{4.52}$$

with the restriction (corresponding to the assumption which has allowed us to omit the inertia in the original momentum equation) that

$$F = \frac{\rho_m}{\rho}\left[0.332\,\frac{L}{R_o}\,R_m^{-1/2} + 0.696\,\frac{L^2}{R_o^2}\,R_m^{-1}\right] >> 1 \tag{4.53}$$

In a typical Wet Spinning process,

$$V_o = 100 \text{ cm/sec} \; ; \; \rho_m = 1.0 \text{ gm/cm}^3 \; ; \; \mu_m = 0.01 \text{ poise} \tag{4.54}$$

Thus, for a spinning way of L = 100cm, it follows from eqs. (4.52) - (4.54) that the minimum viscosity is related to the radius of the spinnerette hole R_o by

$$(\eta_o)_{min} \cong \frac{10}{R_o^2} \text{ poise} \; ; \; R_o \leqslant 0.01 \text{ cm} \tag{4.55}$$

On the other hand, for a fixed value of R_o, typically $R_o = 0.01$ cm, the minimum viscosity is related to the spinning length by

$$(\eta_o)_{min} = 10L^2 \text{ poise} \; ; \; L \geqslant 10 \text{ cm} \tag{4.56}$$

Eqs. (4.55) and (4.56) can be combined to,

$$(\eta_o)_{min} = 10^{-3}\,\frac{L}{R_o} \text{ poise} \; ; \; L \geqslant 10 \text{ cm} \; ; \; \frac{L}{R_o} \geqslant 10^3 \tag{4.57}$$

Inasmuch as $R_o = 0.01$ cm and L = 100 cm (together with the conditions listed in eq. 4.54) are representative values of a typical Wet Spinning process, it follows from eq. (4.57) that a Newtonian fluid can be spun under these conditions only if the viscosity of the fluid is rather high ($> 10^5$ poise).

A filament of a Newtonian fluid of viscosity less than 10^5 poise will, under these conditions, promptly break near the spinnerette hole.

The Full Governing Equation (4.36) and its Solutions

As was pointed out before, a solution to eq. (4.36) can only be obtained numerically. Rather than search for solutions which satisfy a particular value of v_1 at $\xi = 1$, the numerical scheme has been designed to explore the existence of all possible solutions. This can be readily done by varying the value of the velocity gradient at $\xi = 0$, and integrating the momentum equation in the ξ-direction. The curves thus generated exhibit three general properties:

(a) The velocity curves always tend to infinity at a distance which depends on the flow parameters.

(b) For horizontal spinning ($G = 0$), the largest distance along which the velocity remains finite corresponds to the choice of $dv(0)/d\xi = 0$. Further, for the choice of $dv(0)/d\xi > 0$, the velocity curve always lies above the corresponding curve with $dv(0)/d\xi = 0$.

(c) The effect of surface tension is negligibly small in all cases where a solution exists at $\xi = 1$, for $L = O(100\text{cm})$.

In view of properties (a) and (b) we define a limit curve as the velocity curve which remains finite for the largest distance. For horizontal spinning ($G = 0$), the limit curve corresponds, therefore, to the curve with $dv(0)/d\xi = 0$.

The limit curve can be regarded as a bound on existing solutions. This is illustrated below (fig. 4.6) where the shaded area represents the region of all existing solutions. Furthermore, if the limit curve is singular at $\xi < 1$, the boundary condition at $\xi = 1$ can not be satisfied and, hence, no solution exists.

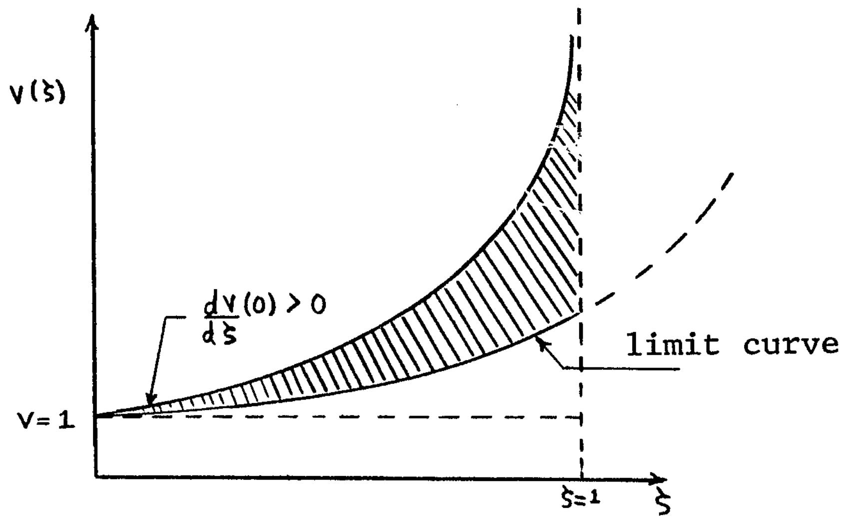

Fig. 4.6 The limit curve.

The limit curves may also represent an estimate of the maximum filament length, L_{max}, that a fluid can form under a prescribed set of conditions. Since the external drag of eq. (4.29) is based, in advance, on a fixed value of L, estimates of the maximum filament length must be regarded as rough approximations. However, for very thin filaments (of the order of 0.01 cm and thinner), the dependance of the external drag on the spinning length L is negligible, and the validity of the above estimates is, therefore, enhanced.

The concept of the limit curve shall be used throughout this study whenever applicable. In order to maintain the two points of view discussed above (i.e. the limit curve as (i) a bound on the region of existing solutions and (ii) an estimate of the maximum spinning length), the limit curves have been plotted in the original dimensional variables.

Figs. (4.7) and (4.8) depict limit curves for the horizontal drawing of filaments of Newtonian fluids. In both of these figures, the spinning conditions correspond to a typical Dry Spinning process with air as the surrounding medium ($\rho_m = 1.2 \cdot 10^{-3}$gm/cm^3, $\mu_m = 2 \cdot 10^{-4}$ poise). The external drag has been based on L = 100 cm. Figs. (4.7) and (4.8) (corresponding, respectively, to values of R_o of 0.1 and 0.01 cm) show the variations in the limit curves for several values of the filament viscosity. The latter is represented in terms of the Trouton viscosity η_T which, for a Newtonian fluid, is related to η_o by

$$\eta_T = 3\eta_o$$

From these figures, it is possible again to estimate the minimum viscosity which is required for the formation of a filament, 100 cm long, under the conditions shown. Accordingly,

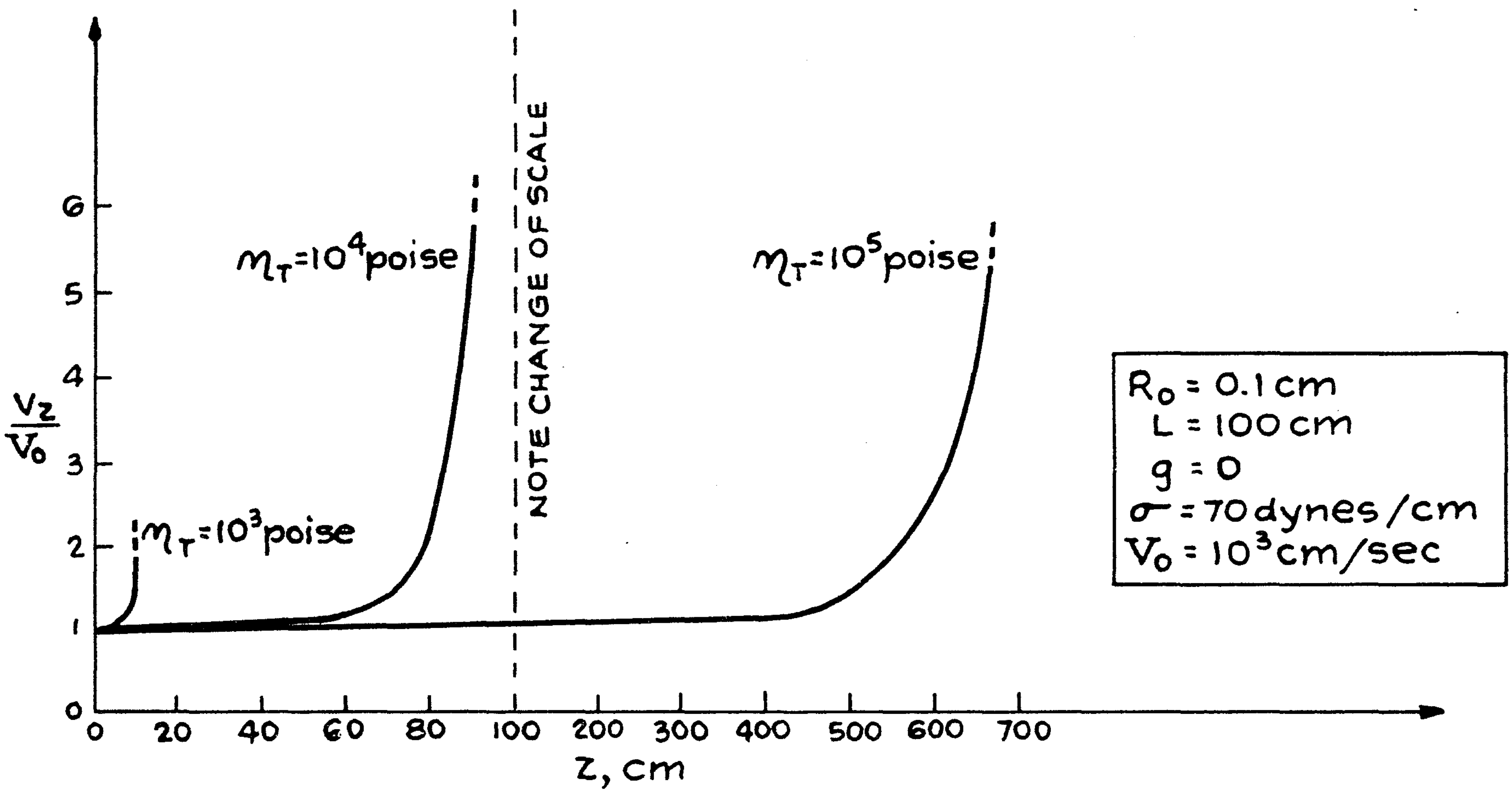

FIG. 4.7 LIMIT CURVES FOR A NEWTONIAN FLUID.

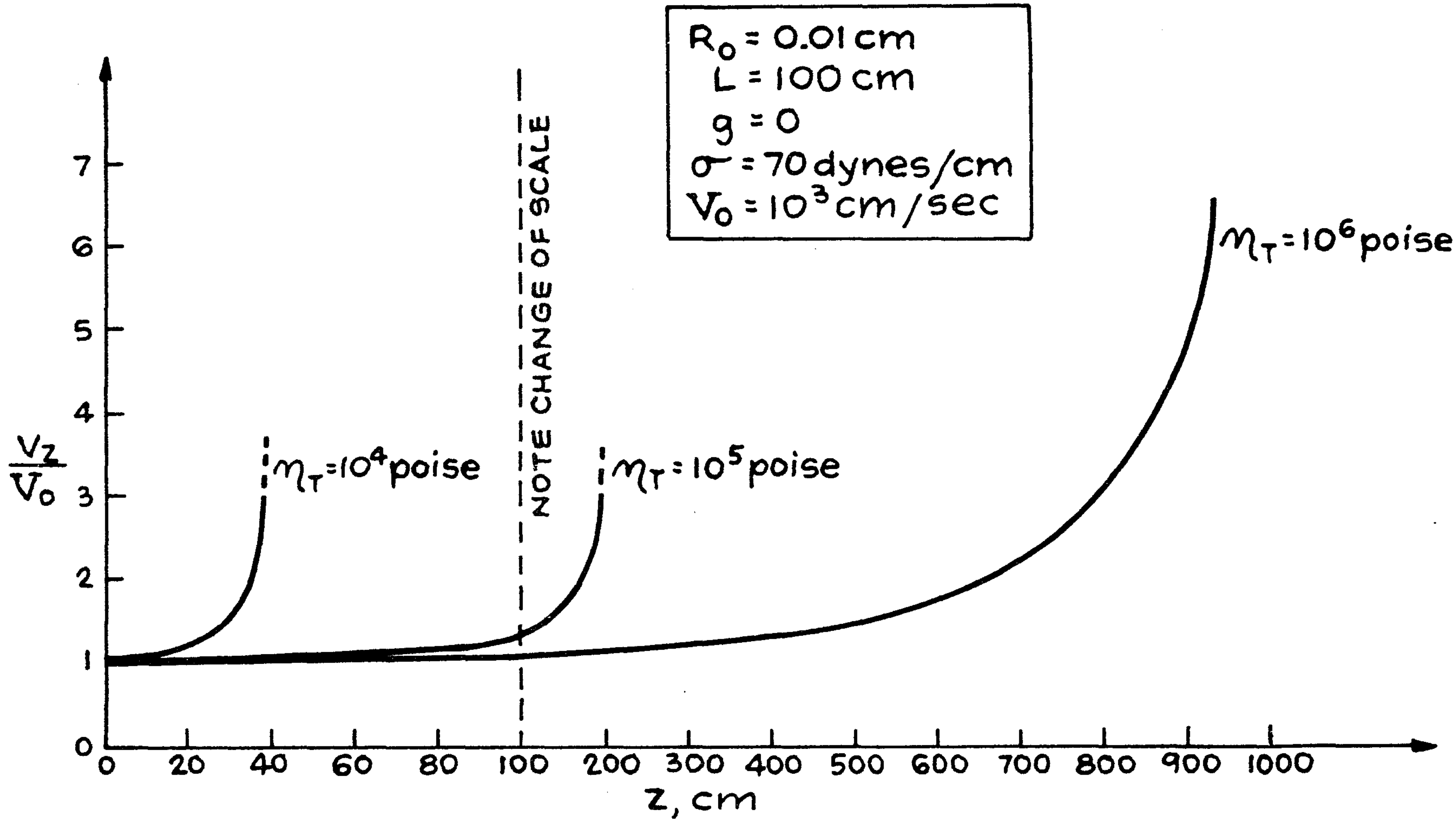

FIG. 4.8 LIMIT CURVES FOR A NEWTONIAN FLUID.

for $R_o = 0.1$ cm, $(\eta_o)_{min} = 4 \cdot 10^3$ poise

and

for $R_o = 0.01$ cm, $(\eta_o)_{min} = 2 \cdot 10^4$ poise

For values of R_o appreciably smaller than 0.01 cm, the inertia becomes negligibly small compared to the drag, and the minimum viscosity may be estimated with the aid of eq. (4.52) (corresponding to the solution without inertia). Thus,

for $R_o = 0.001$ cm, $(\eta_o)_{min} = 3\ 10^5$ poise

The above estimates are summarized below:

R_o; cm	$(\eta_o)_{min}$; poise
0.1	$4 \cdot 10^3$
0.01	$2 \cdot 10^4$
0.001	$3 \cdot 10^5$

Table 4.1 Minimum viscosity as a function of the spinnerette hole radius, R_o, for Dry Spinning (conditions are shown in figs. 4.7 and 4.8).

The Effect of Gravity

In many of the commercial spinning processes, the filaments are drawn vertically downwards. Such an arrangement may be due to technical convenience since, otherwise, the effect of gravity is not too appreciable. On the whole, the effect of gravity is favorable to spinning since the necessary force applied at the wind-up roll (corresponding to a given pullaway velocity), is smaller when the filaments are drawn downwards than in the case in which the filaments are drawn horizontally. However, on the basis of our results, it is doubtful that one can form a satisfactory fiber in "gravitational spinning" if a similar attempt has failed with the drawing maintained horizontally. Fig. 4.9 depicts two limit curves corresponding to gravitational and horizontal spinning.

Note that when the effect of gravity is included, the limit curve does not necessarily correspond to the solution of eq. (4.36) with $dv(0)/d\xi = 0$. Rewriting eq. (4.36) in the form

$$\frac{d^2v}{d\xi^2} = R_L v\frac{dv}{d\xi} + \frac{1}{v}\left(\frac{dv}{d\xi}\right)^2 + 2FR_L v^2 - G \qquad (4.58)$$

it can be seen that if

$$dv(0)/d\xi = 0 \ ; \quad 2FR_L < G \qquad (4.59)$$

it follows that

$$d^2v(0)/d\xi^2 < 0 \qquad (4.60)$$

Consequently, the velocity will decrease along some portion or, all along the spinning way; a situation which does not conform to reality. Therefore, when the inequality of (4.59) holds, the limit curve will correspond to a solution of eq. (4.36) with $dv(0)/d\xi > 0$.

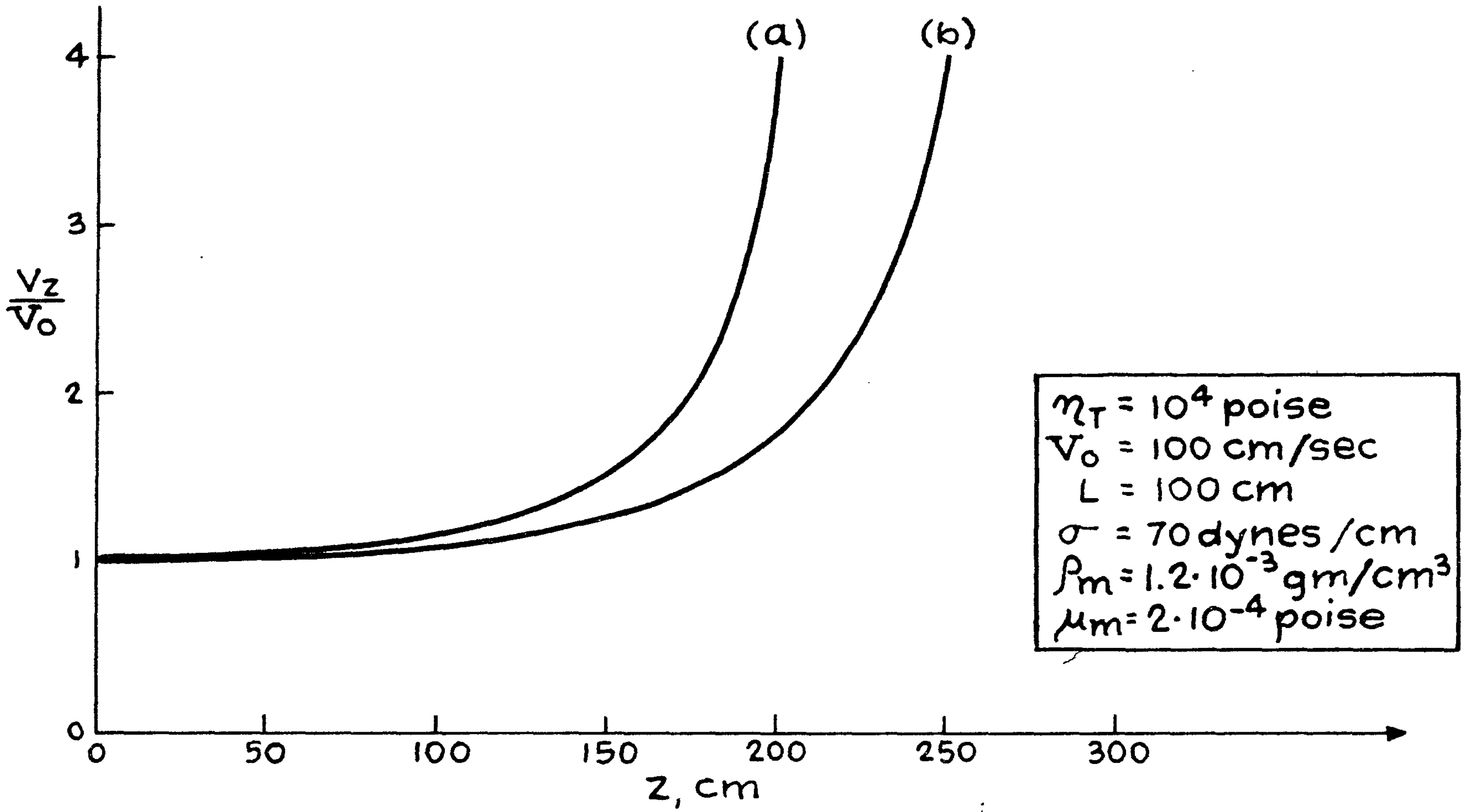

FIG. 4.9 THE EFFECT OF GRAVITY UPON THE SPINNING OF A NEWTONIAN FLUID. LIMIT CURVES: (a) WITHOUT GRAVITY (b) WITH GRAVITY.

<u>The Effect of Hardening</u>

The formation of self-supporting fibers of sufficient strength is generally accompanied by a viscosity increase of several orders of magnitude. This hardening process is caused by the exchange of mass or heat (depending on the particular spinning method) between the filament and its surrounding. A proper description of the hardening phenomenon must involve the simultaneous application of energy, mass and momentum balances, a problem which at present can not be solved. In addition, the knowledge of the dependence of the material properties on heat and mass transfer is quite inadequate. Therefore, in order to ascertain the effect of hardening, the viscosity increase along the spinning way has been described in an arbitrary fashion (disregarding mass and energy changes) by an expression such as

$$\eta(z) = \eta_o(1 + mz^2) \tag{4.61a}$$

Or, in a dimensionless form

$$R_L(z) = R_{Lo}(1 + M\ ^2)^{-1} \; ; \; M = mL \tag{4.61b}$$

where η_o and R_{L_o} are the initial values of the viscosity and the Reynolds number at the exit nozzle.

Fig. 4.10 compares the spinning of a filament of a Newtonian fluid with and without hardening. Eq. (4.61), with m = 0.5 (or, equivalently, M = 50 for L = 100 cm), has

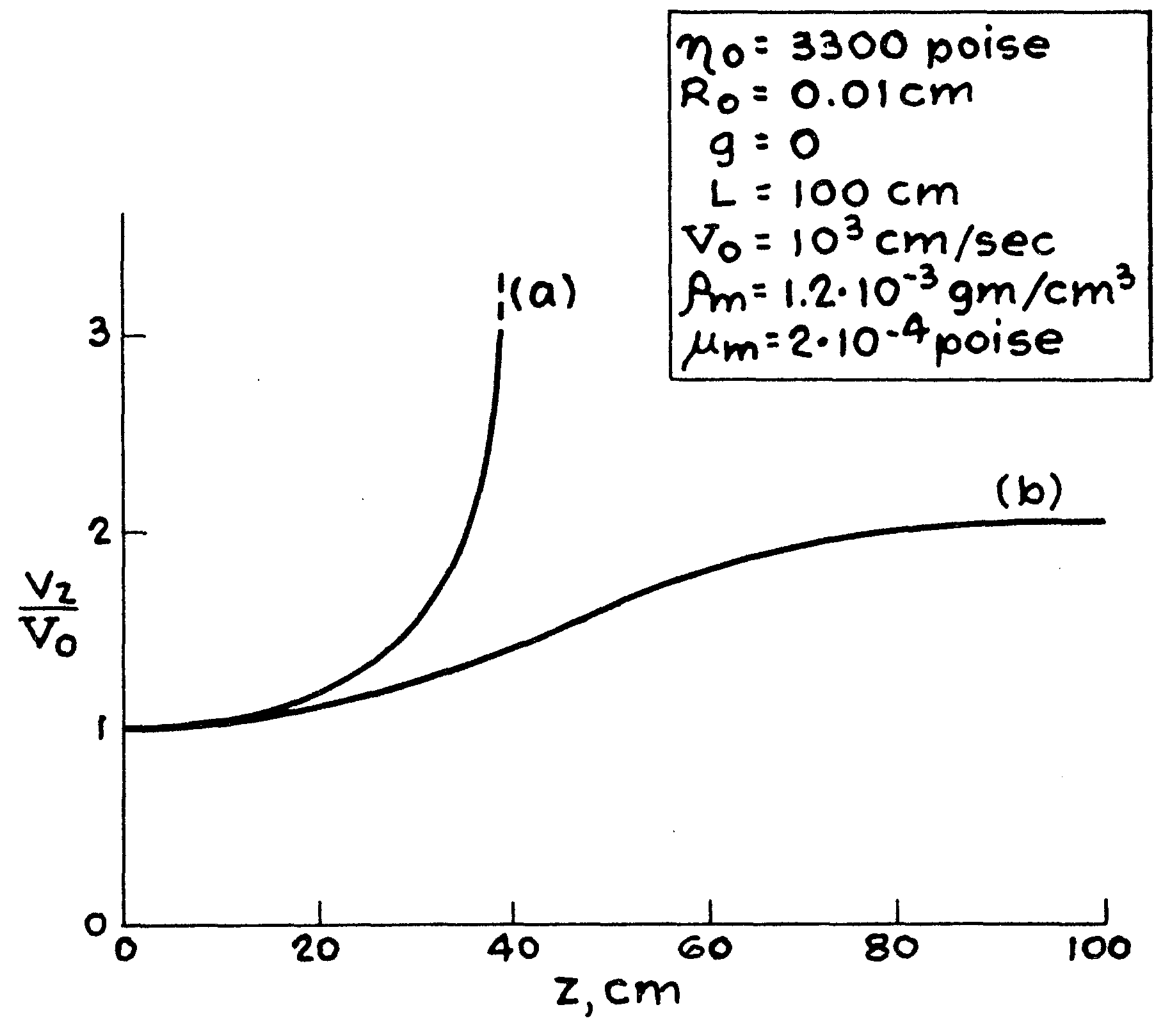

FIG. 4.10 LIMIT CURVES FOR A NEWTONIAN FLUID, (a) WITHOUT HARDENING, (b) WITH HARDENING.

been used to describe the viscosity increase along the spinning way due to hardening. Fig. 4.10 makes it clear that a filament which otherwise would break up (no hardening), can be readily spun when the effect of hardening is appreciable (corresponding in the present case to a fiftyfold increase in the viscosity along a spinning way of 100 cm). Note, further, the occurence of an inflexion point in the velocity curve, the presence of which is typical to all industrial spinning data.

4.5 SOLUTIONS FOR FILAMENTS OF NON-NEWTONIAN FLUIDS

When one enters the domain of non-Newtonian fluids, the first difficulty which arises is that of selecting a suitable constitutive equation which, together with the momentum equation, can furnish a solution subject to the specified boundary conditions. Thus, pursuing the direct approach, we find that the more general constitutive equations such as Noll's simple fluid theory, are mathematically unmanageable in the present case. This, even in view of the simplifying assumption that the axial velocity, v_z, is independent of the radial direction. Instead, one must resort to any of the relatively simple, explicit, constitutive equations which have been, hitherto, used by rheologists with some degree of success. Accordingly, three equations of this type have been selected. The first, representing a viscoinelastic fluid, is the Reiner-Rivlin constitutive equation which has the form

$$\underset{\sim}{T} = 2\beta_0 \underset{\sim}{D} + 4\beta_1 \underset{\sim}{D}^2 \tag{4.62}$$

where $\underset{\sim}{D}$ is the rate of strain tensor, and β_0 and β_1 are positive material constants. The two other equations represent two different non-linear generalizations of the linear Maxwell fluid: The first

$$\underset{\sim}{T} + \lambda \frac{d_c \underset{\sim}{T}}{dt} = 2\eta_0 \underset{\sim}{D} \tag{4.63}$$

is commonly referred to as <u>Oldroyd fluid A</u>, and

$$\underset{\sim}{T} + \lambda \frac{d^c \underset{\sim}{T}}{dt} = 2\eta_0 \underset{\sim}{D} \tag{4.64}$$

is commonly called the <u>Oldroyd fluid B</u>. In eqs. (4.63) and (4.64) η_0 is the zero viscosity, λ is a positive fluid constant and d_c/dt and d^c/dt are the convected derivatives defined by eqs. (1.57) and (1.58).

<u>Reiner-Rivlin Fluid</u>

In accordance with the simplifying assumptions of eq. (4.14), the stress distribution in a filament of a Reiner-Rivlin fluid is given by

$$T_{rr} = -\beta_0 \frac{dV_z}{dz} + \beta_1 \left(\frac{dV_z}{dz}\right)^2 \tag{4.65}$$

$$T_{zz} = 2\beta_0 \frac{dV_z}{dz} + 4\beta_1 \left(\frac{dV_z}{dz}\right)^2 \tag{4.66}$$

It follows that

$$T_{zz} - T_{rr} = 3\beta_0 \frac{dV_z}{dz} + 3\beta_1\left(\frac{dV_z}{dz}\right)^2 \tag{4.67}$$

or

$$T_{zz} - T_{rr} = 3\beta_0 \frac{dV_z}{dz}\left(1 + \lambda_2 \frac{dV_z}{dz}\right) \tag{4.68}$$

where

$$\lambda_2 = \beta_1/\beta_0$$

In the absence of surface tension effects

$$T_{zz} - T_{rr} = S_{zz}$$

so that eq. (4.68) may be used to define the elongational, or Trouton viscosity, for a Reiner-Rivlin fluid, i.e.,

$$(\eta_T)_{R.L.} = \frac{S_{zz}}{(dV_z/dz)} = 3\beta_0\left(1 + \lambda_2 \frac{dV_z}{dz}\right) \tag{4.69}$$

where the initials R.L. serve to distinguish $(\eta_T)_{R.L.}$ from the corresponding notation for a Newtonian fluid.

Substituting eq. (4.68) into eq. (4.30), and casting the resulting equation in a dimensionless form, yields

$$R_L \frac{dV}{d\xi} = \frac{d}{d\xi}\left\{\frac{1}{V}\left[\frac{dV}{d\xi} + \beth\left(\frac{dV}{d\xi}\right)^2\right]\right\} - 2FR_L V + \frac{G}{V} \tag{4.70}$$

where

$$R_L = \frac{\rho V_0 L}{3\beta_0}, \quad \beth = \frac{\lambda_2 V_0}{L}$$

The boundary conditions remain

$$v(0) = 1 \ ; \ v(1) = v_1 \ ; \ v_1 > 1 \tag{4.71}$$

When $\beth = 0$ ($\lambda_2 = 0$), eq. (4.70) clearly becomes identical to the corresponding equation for a filament of a Newtonian fluid (4.36). A comparison between the spinnability of a filament of a Reiner-Rivlin fluid and that of a Newtonian fluid can, therefore, be drawn on the basis of an equivalent zero viscosity ($\eta_o = \beta_o$). Figs. 4.11 and 4.12 provide such a comparison. The spinning conditions are those typical to Dry Spinning processes with air as the surrounding medium ($\rho_m = 1.2 \cdot 10^{-3}$ gm/cm^3, $\mu_m = 2 \cdot 10^{-4}$ poise). The values of the zero viscosities which have been selected are $3\eta_o = 10^4$ poise (for fig. 4.11) and $3\eta_o = 10^3$ (for fig. 4.12). The values of λ_2 have been varied of several decades of magnitude. The solutions shown in these figures are the limit curves corresponding to the solutions of the momentum equation with $dv(0)/d\xi = 0$.

From figs. 4.11 and 4.12 it is obvious that Newtonian fluids (with values of η_o given above) can not be spun over a spinning length of 100 cm, whereas a Reiner-Rivlin fluid can readily be spun over this length provided that λ_2 is sufficiently large. In other words, if the apparent viscosity increases sufficiently quickly with the rate of elongation,

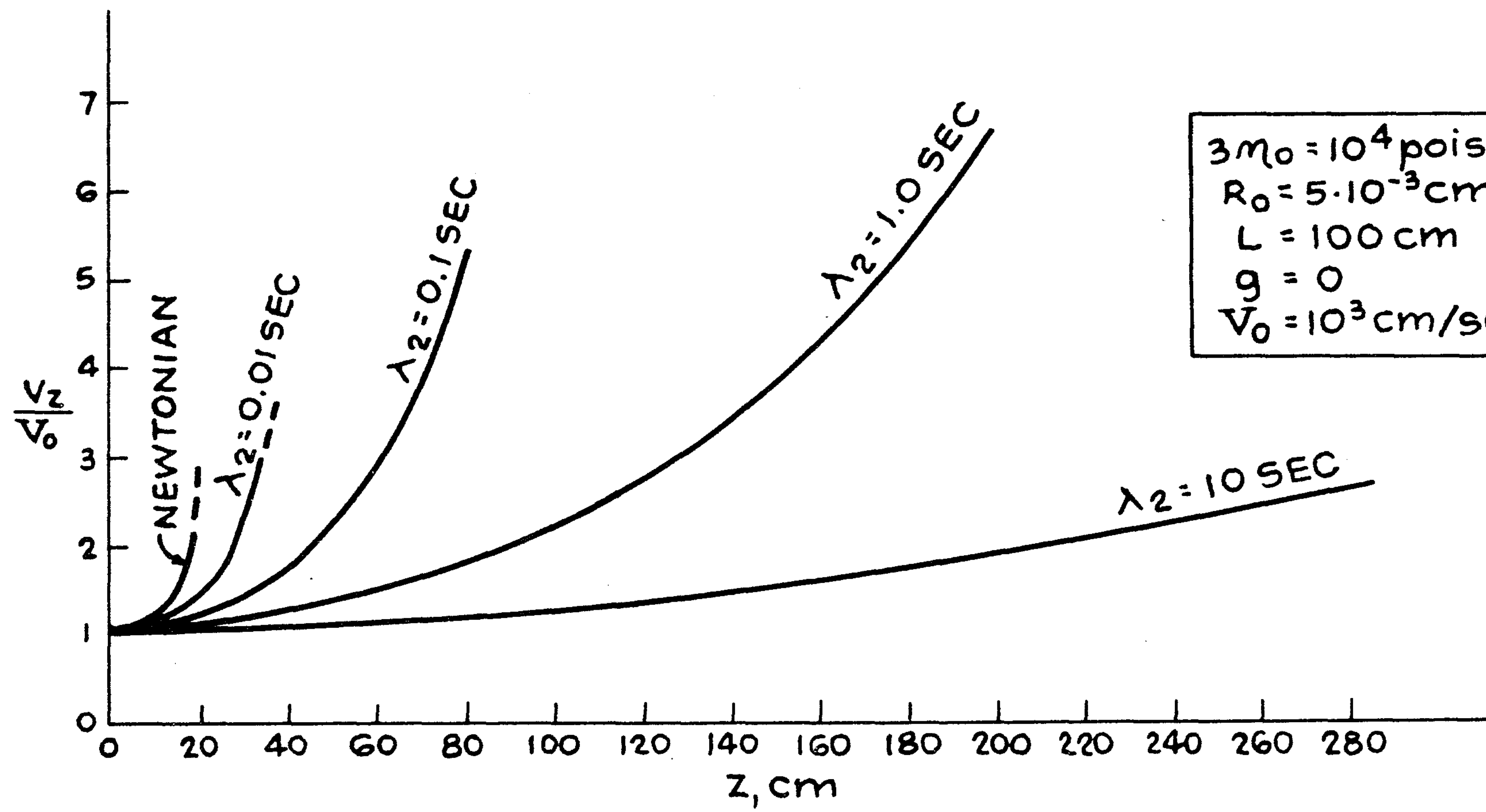

FIG. 4.11 COMPARISON BETWEEN THE LIMIT CURVES FOR FILAMENTS OF A NEWTONIAN AND REINER-RIVLIN FLUIDS.

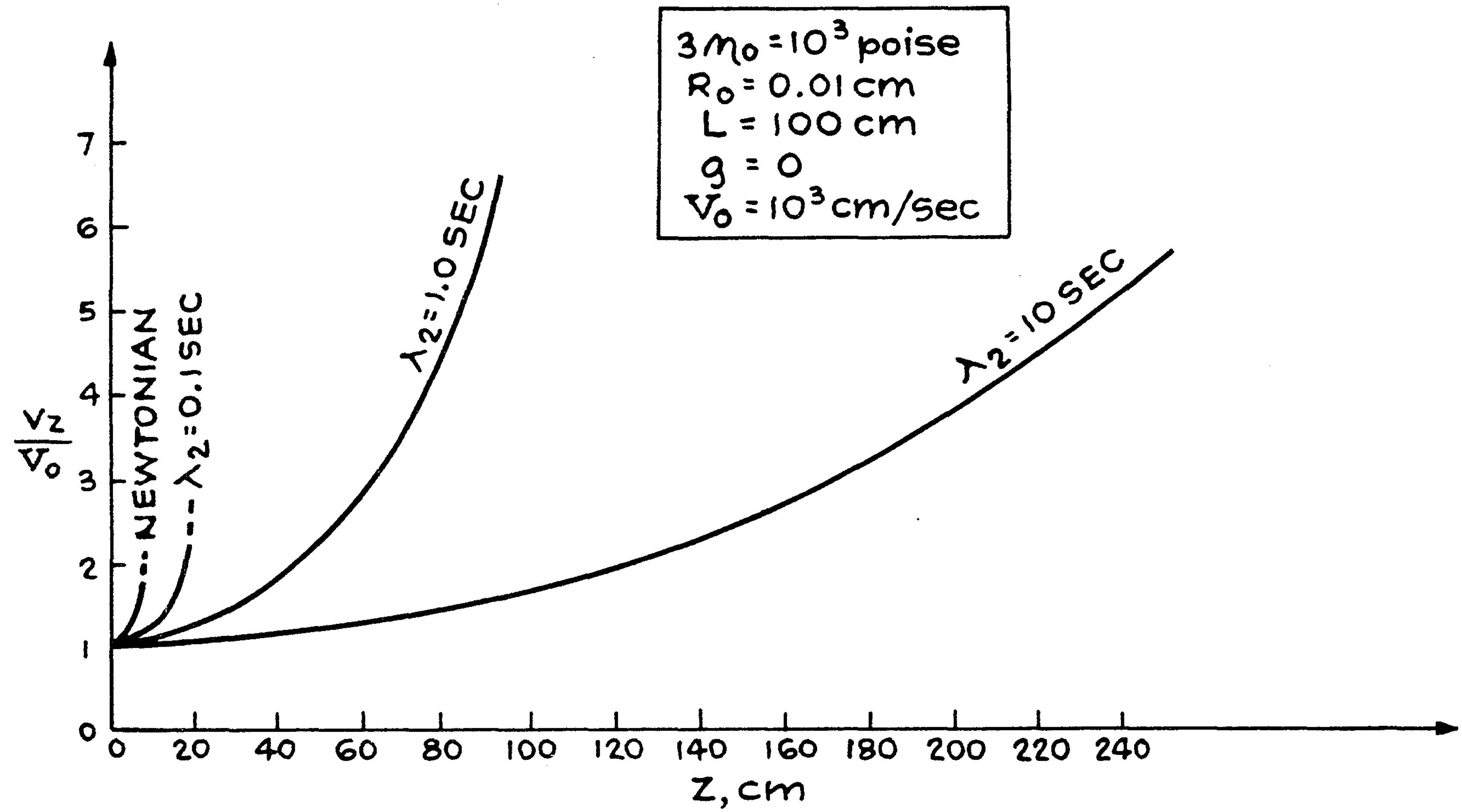

FIG. 4.12 COMPARISON BETWEEN THE LIMIT CURVES FOR FILAMENTS OF A NEWTONIAN AND REINER-RIVLIN FLUIDS.

a filament of a fluid with a relatively small zero viscosity can be spun without hardening.

Reference to eq. (4.69) shows indeed that, for any positive value of λ_2, the elongational viscosity of a Reiner-Rivlin fluid increases with the elongation rate.

Oldroyd Fluids A and B

The stress distribution in filaments of Oldroyd fluids A and B is given by:

Oldroyd fluid A,

$$T_{rr}\left(1 - \lambda \frac{dv_z}{dz}\right) + \lambda v_z \frac{dT_{rr}}{dz} = -\eta_0 \frac{dv_z}{dz} \qquad (4.72a)$$

$$T_{zz}\left(1 + 2\lambda \frac{dv_z}{dz}\right) + \lambda v_z \frac{dT_{zz}}{dz} = 2\eta_0 \frac{dv_z}{dz} \qquad (4.72b)$$

Oldroyd fluid B,

$$T_{rr}\left(1 + \lambda \frac{dv_z}{dz}\right) + \lambda v_z \frac{dT_{rr}}{dz} = -\eta_0 \frac{dv_z}{dz} \qquad (4.73a)$$

$$T_{zz}\left(1 - 2\lambda \frac{dv_z}{dz}\right) + \lambda v_z \frac{dT_{zz}}{dz} = 2\eta_0 \frac{dv_z}{dz} \qquad (4.73b)$$

Unlike the previous cases, the use of the above equations to eliminate T_{rr} and T_{zz} appearing in the momentum equation (4.30) can not be accomplished. Instead, it is necessary to obtain three first-order differential equations of the form

$$dv_z/dz = f(v_z, T_{zz}, T_{rr})$$

$$dT_{rr}/dz = g(v_z, T_{zz}, T_{rr}) \qquad (4.74)$$

$$dT_{zz}/dz = h(v_z, T_{zz}, T_{rr})$$

The resulting equations are rather lengthy; cast in a dimensionless form they are:

Oldroyd fluid A,

$$\frac{dV}{d\xi} = \frac{2FR_L\lambda V^3 + T_Z - T_R}{3 - 3\lambda T_Z - \lambda R_L V^2} \tag{4.75a}$$

$$\frac{dT_Z}{d\xi} = \frac{4FR_L(1-\lambda T_Z) + R_L V T_Z + (T_Z^2 - T_Z/\lambda - 2T_R/\lambda + 2T_Z T_R)/V}{3 - 3\lambda T_Z - \lambda R_L V^2} \tag{4.75b}$$

$$\frac{dT_R}{d\xi} = \frac{2FR_L\lambda V^3(\lambda T_R - 1) - (T_Z - T_R) - T_R(3 - 3\lambda T_Z - \lambda R_L V^2)}{(3 - 3\lambda T_Z - \lambda R_L V^2)\lambda V} \tag{4.75c}$$

Oldroyd fluid B

$$\frac{dV}{d\xi} = \frac{2FR_L\lambda V^3 + T_Z - T_R}{3 - \lambda R_L V^2 + \lambda T_Z + 2\lambda T_R} \tag{4.76a}$$

$$\frac{dT_Z}{d\xi} = \frac{4FR_L V^2(1+\lambda T_Z) - 2T_R(1+\lambda T_Z)/(\lambda V) + T_Z(R_L V + T_Z/V - 2T_R/V - 1/\lambda V)}{3 - \lambda R_L V^2 + \lambda T_Z + 2\lambda T_R} \tag{4.76b}$$

$$\frac{dT_R}{d\xi} = -\frac{(1+\lambda T_R)(2FR_L\lambda V^3 + T_Z - T_R) - T_R(3 - \lambda R_L V^2 + \lambda T_Z + 2\lambda T_R)}{(3 - \lambda R_L V^2 + \lambda T_Z + 2\lambda T_R)\lambda V} \tag{4.76c}$$

where

$$R_L = \frac{\rho V_0 L}{3\eta_0}, \quad T_Z = \frac{T_{zz}}{\eta_0 V_0/L}, \quad T_R = \frac{T_{rr}}{\eta_0 V_0/L}, \quad \lambda = \frac{\lambda V_0}{L} \tag{4.77}$$

The above equations, admittedly, are very complex and a mere visual examination does not seem to furnish any significant information concerning the spinning of fluids obeying these equations. The numerical results show that there is no appreciable difference in the results obtained for fluids A and B. This is in marked contrast to steady shear flow in which the constitutive equations for these two fluids give rise to fundamentally different results.

In order to obtain numerical results it is first necessary to select the initial values of T_z and T_R (i.e., $T_z(0)$ and $T_R(0)$). Several suitable selections have been tried and it appears that the choice

$$T_z(0) = 0 \ , \quad T_R(0) = 0 \tag{4.78}$$

provides the limit curves of the solutions in the sense that $v(\xi)$ described by these curves remains finite for the largest distance. The resulting solutions show that both fluids A and B are <u>less</u> spinnable than a Newtonian fluid of the same zero viscosity. That is, as λ increases the maximum filament length (corresponding to the limit curves) decreases. This is illustrated in fig. 4.13 in which the limit curves for fluid A, corresponding to different values of λ, are shown. Since λ represents a measure of the elasticity of the fluid, the above results stand in contradiction to the physical evidence which indicates that viscoelastic fluids are appreciably more spinnable than Newtonian fluids.

In order to understand the reason for this discrepancy

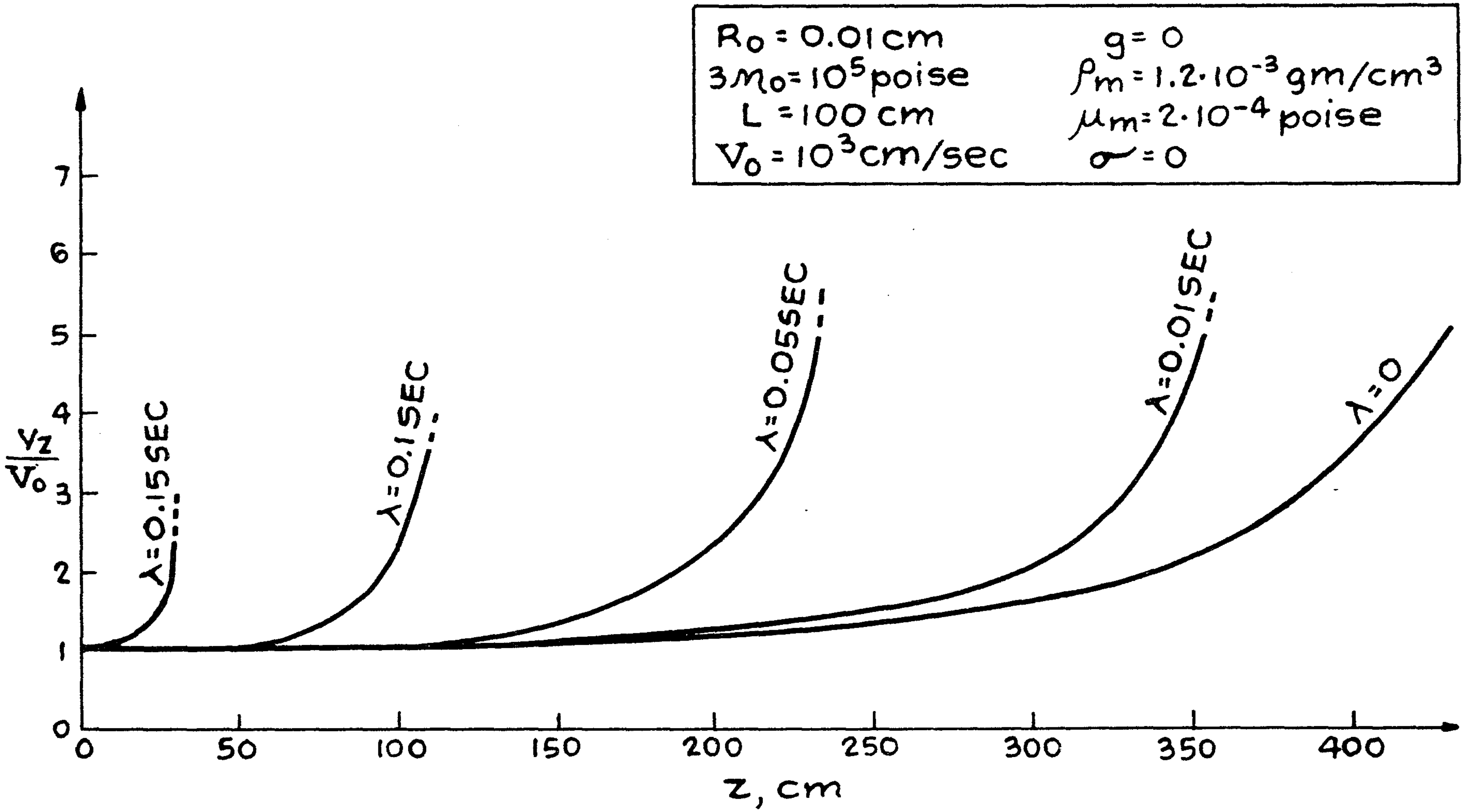

FIG. 4.13 LIMIT CURVES FOR OLDROYD FLUID A.

consider the stress distribution in a filament of an Oldroyd fluid A given by eqs. (4.72a) and (4.72b). Denoting dv_z/dz, dT_{rr}/dz and dT_{zz}/dz by v_z' , T_{rr}' and T_{zz}', respectively, eqs. (4.72a) and (4.72b) can be written as

$$T_{rr} = -\frac{\eta_0 V_z' - \lambda V_z T_{rr}'}{1-\lambda V_z'} \tag{4.79a}$$

$$T_{zz} = \frac{2\eta_0 V_z - \lambda V_z T_{zz}'}{1 + 2\lambda V_z'} \tag{4.79b}$$

From which (in the absence of surface tension)

$$S_{zz} = T_{zz} - T_{rr} = \frac{3\eta_0 V_z' - \lambda V_z T_{zz}'(1-\lambda V_z') + \lambda V_z T_{rr}'(1+2\lambda V_z')}{(1-\lambda V_z')(1+2\lambda V_z')} \tag{4.80}$$

Consider now the case in which the velocity gradient increases and approaches the value

$$V_z' = \frac{1}{\lambda} \tag{4.81}$$

In this case, it follows from eq. (4.72a) that

$$T_{rr}' = -\frac{\eta_0 V_z'}{\lambda V_z} = -\frac{\eta_0}{\lambda^2 V_z} \tag{4.82}$$

Eq. (4.80) thus becomes

$$S_{zz} = \frac{2\eta_0 V_z'}{1+2\lambda V_z'} = \frac{2}{3\lambda}\eta_0 \tag{4.83}$$

and the elongational viscosity $(\eta_T)_A$ is

$$(\eta_T)_A = \frac{S_{zz}}{V_z'} = \frac{2}{3}\eta_0 \tag{4.84}$$

Thus as the velocity gradient increases, the elongational viscosity, $(\eta_T)_A$, of the Oldroyd fluid A decreases. As v_z' approaches the value of $1/\lambda$, the elongational viscosity becomes $(2/3)\eta_o$ which is of course smaller than the elongational viscosity of a Newtonian fluid for which

$$\eta_T = 3\eta_o$$

For an Oldroyd fluid B, it follows from eqs. (4.73a) and (4.73b) that

$$S_{zz} = T_{zz} - T_{rr} = \frac{3\eta_o V_z' - \lambda V_z T_{zz}'(1+\lambda V_z') + \lambda V_z T_{rr}'(1-2\lambda V_z')}{(1+\lambda V_z')(1-2\lambda V_z')} \qquad (4.85)$$

In this case, as v_z' increases and approaches the value

$$V_z' = \frac{1}{2\lambda} \qquad (4.86)$$

it follows from eq. (4.73b) that

$$T_{zz}' = \frac{2\eta_o V_z'}{\lambda V_z} = \frac{\eta_o}{\lambda^2 V_z} \qquad (4.87)$$

and, hence, S_{zz} becomes

$$S_{zz} = \frac{\eta_o V_z'}{1+\lambda V_z'} = \frac{2\eta_o}{3\cdot 2\lambda} \qquad (4.88)$$

Consequently, when $v_z' = 1/2$ the elongational viscosity, $(\eta_T)_B$ of the Oldroyd fluid B is

$$(\eta_T)_B = \frac{2}{3}\eta_o \qquad (4.89)$$

Here again, as the velocity gradient increases, the resistance of the fluid to streching (represented by the elongational viscosity) decreases. Based on the forgoing analysis, therefore, it appears that the constitutive equations of the Oldroyd fluids are unsuitable for the description of the spinning of viscoelastic fluids.

The above conclusion stands in opposition to several works in which the constitutive equations of the Oldroyd type are claimed to be proper representatives of viscoelastic fluids in spinning processes. Metzner [37], Pearson [47] and others have shown that the constitutive equations of the Oldroyd type do predict an elongational viscosity which increases with the elongational rate. However, the works cited above are confined only to the study of steady extension with negligible inertia. In steady extension the velocity gradient is a constant, $dv_z/dz = e$ say; consequently, it is possible to show that, in the absence of inertia and surface effects, the stresses T_{zz} and T_{rr} are both constants. Under these circumstances, the stresses in a filament of an Oldroyd fluid B, for example, can be obtained from eqs. (4.73a) and (4.73b) by letting

$$\text{(i) } dv_z/dz = \text{constant} = e \qquad (4.90)$$

$$\text{(ii) } T'_{zz} = T'_{rr} = 0 \qquad (4.91)$$

Accordingly,

$$T_{rr} = -\frac{\eta_0 e}{1+\lambda e} \qquad (4.92)$$

$$T_{zz} = \frac{2\eta_0 e}{1-2\lambda e} \qquad (4.93)$$

so that S_{zz}, the total tensile stress, is

$$S_{zz} = \frac{3\eta_0 e}{(1+\lambda e)(1-2\lambda e)} \qquad (4.94)$$

and the elongational viscosity is

$$(\eta_T)_B = \frac{3\eta_0}{(1+\lambda e)(1-2\lambda e)} \qquad (4.95)$$

Eq. (4.95) indeed predicts that as the rate of elongation "e" increases, the elongational viscosity increases as well. Note, however, that when the velocity gradient is not regarded as a constant, as is the case in the present work, the omission of the stress gradient terms T'_{zz} and T'_{rr} is no longer justified. In fact, in the absence of inertia and surface effects, the momentum equation of (4.30) reduces to,

$$\frac{d}{dz}\left[\frac{1}{V_z}(T_{zz} - T_{rr})\right] = 0 \qquad (4.96)$$

from which

$$V_z(T'_{zz} - T'_{rr}) = V'_z(T_{zz} - T_{rr}) \qquad (4.97)$$

From eq. (4.97) it is clear that the terms in eqs. (4.73a) and (4.73b) which involve the stress gradients T'_{zz} and T'_{rr} are of comparable magnitude to the other terms and, hence, can not be omitted (as is the case in steady extension). Steady extension, indeed, constitutes a highly specialized description of a filament under tension. Consequently, analogies between steady extension and the behavior of liquid filaments in spinning processes are not necessarily obvious. Thus, whereas in steady extension fluids of the Oldroyd type can exhibit an increasing elongational viscosity, in the more general case considered in the present section, the Oldroyd fluids exhibit the opposite effect and, hence, can not be regarded as proper representatives of viscoelastic fluids in spinning process. A word of qualified reservation should, however, be added: Since the present analysis is based on the simplifying assumptions of (4.14), it is possible that a more rigorous analysis might lead to different results. However, inasmuch as the present analysis constitutes a reasonable approximation to a more general solution, the latter alternative does not seem very likely.

4.6 DISCUSSION

The analysis of the mechanics of spinning, presented in the forgoing pages, is based upon constitutive equations which describe fluids. This is not to say that the materials which are processed industrially to form fibers are real fluids. Indeed, under actual spinning conditions the filament develops structural anisotropy and exhibits solid-like characteristics [56]. However, the principal aim of this study has been to examine the limiting case in which energy and mass transfer may be ignored and, under these circumstances, to ascertain the effect of the inherent rheological properties of fluids upon their spinnability. Other considerations, which admittedly are very valid, are beyond the scope of this study.

Turning first to the case in which the filament fluid is Newtonian. The results of the preceding analysis show that, under typical spinning conditions, a Newtonian fluid is spinnable only if its viscosity is rather high, i.e., of the order of 10^5 poise. However, it is very doubtful that such a viscous fluid can be forced out of the narrow spinnerette holes without the application of an enormous pressure difference. For example, in order to maintain an average extrusion speed of 100 cm/sec through a spinnerette hole of 10^{-2} cm in radius, the necessary applied pressure should be of the order of 10^7 psi per 1 cm of nozzle length. It seems, therefore, that without hardening the spinning of a Newtonian fluid is not too realistic.

On the other hand, the mechanism of hardening makes it possible to employ Newtonian fluids of viscosities which are considerably less than 10^5 poise. In this case the fluid is extruded at elevated temperatures or diluted in a suitable volatile solvent. The resulting filaments subsequently attain high viscosities due, depending on the spinning method, to the fast cooling of the filaments, or due to the coagulation mechanism. Under these circumstances, a Newtonian fluid may form long filaments; this has been illustrated in fig. 4.10 in which the viscosity increase due to hardening has been described empirically. Indeed, it has been reported that some polymers which are generally melt-spun exhibit very small deviations from a Newtonian behavior [21, 35].

What are the effects of the non-Newtonian properties on spinnability? It has been postulated before, and it also appears from the present analysis that the dominant property of non-Newtonian fluids which account for their superior spinnability is the increase of the elongational viscosity with the extension rate. Fluids for which the elongational viscosity increases strongly enough with the extension rate should, therefore, be capable of producing filaments without hardening. In addition, non-Newtonian fluids are more suitable to the spinning process because of their shear thinning property (decrease in the apparent viscosity with the shear rate). This property makes it possible to extrude these fluids at high speeds through the narrow holes of the spinnerette without the application of an excessive pressure difference.

Three non-Newtonian fluids have been considered in this work: The Reiner-Rivlin fluid, representing a viscoinelastic fluid, and the two Oldroyd fluids which are non-linear generalizations of the linear Maxwell fluid. Among these three, only the Reiner-Rivlin fluid exhibits an elongational viscosity which increases with the rate of elongation. The two Oldroyd fluids exhibit the opposite effect. Thus, a Reiner-Rivlin fluid of relatively small zero viscosity may form long filaments without hardening provided that the increase in the elongational viscosity with the elongation rate is sufficiently strong (a large β_1 in eq. 4.62). On the other hand, fluids obeying the constitutive equations of (4.63) and (4.64) (corresponding, respectively, to Oldroyd fluids A and B) are even less spinnable than a Newtonian fluid of the same zero viscosity. Since this is contradictory to the physical evidence, the constitutive equations of the Oldroyd fluids should not be regarded as proper representatives of viscoelastic fluids in spinning processes.

Let us now examine typical spinning data. Fig. 4.14 shows the variation of the radius of a filament of Nylon 6 formed in Melt Spinning as reported by Mark [33]. Fig. 4.15 shows the corresponding axial velocity variation along the spinning way. The velocity profile of fig. 4.15 has been obtained by means of the continuity equation (4.17). One characteristic feature of these curves stands out, namely, that both velocity and the filament radius become nearly uniform some finite distance away from the wind-up roll. In consequence, the velocity curve exhibits an inflexion point. The same pattern exists in all the spinning data which has been examined by the author. Now, there is no doubt that the spinning behavior exemplified in figs. 4.14 and 4.15 is largely due to the enormous increase in viscosity due to hardening. Indeed, the region in which the velocity becomes nearly uniform corresponds to a viscosity range where the filament is, for all practical purposes, a solid. Our analysis of the spinning of a Newtonian fluid, with the effect of hardening included, also gave rise to a velocity curve with a point of inflexion. The question is, since viscoelastic fluids appear to posses the property whereby their apparent viscosity increases with the strain rate, is it possible that the type of behavior shown in figs. 4.14 and 4.15

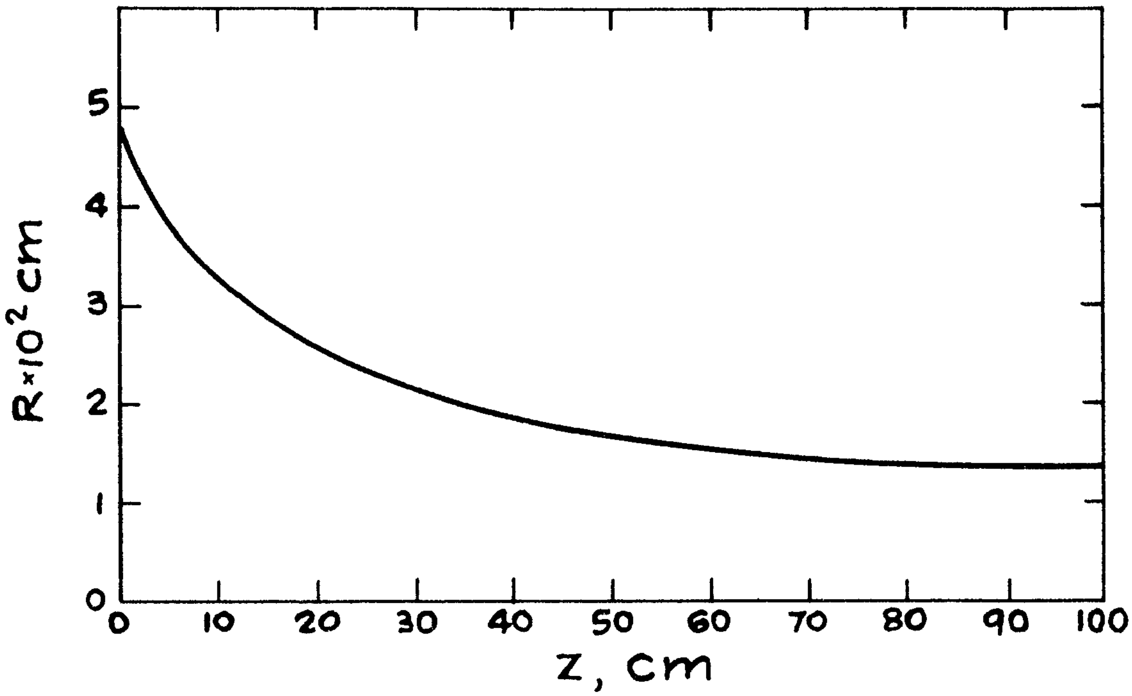

FIG. 4.14 THE VARIATION OF THE FILAMENT RADIUS ALONG THE SPINNING WAY IN THE SPINNING OF NYLON 6 [33].

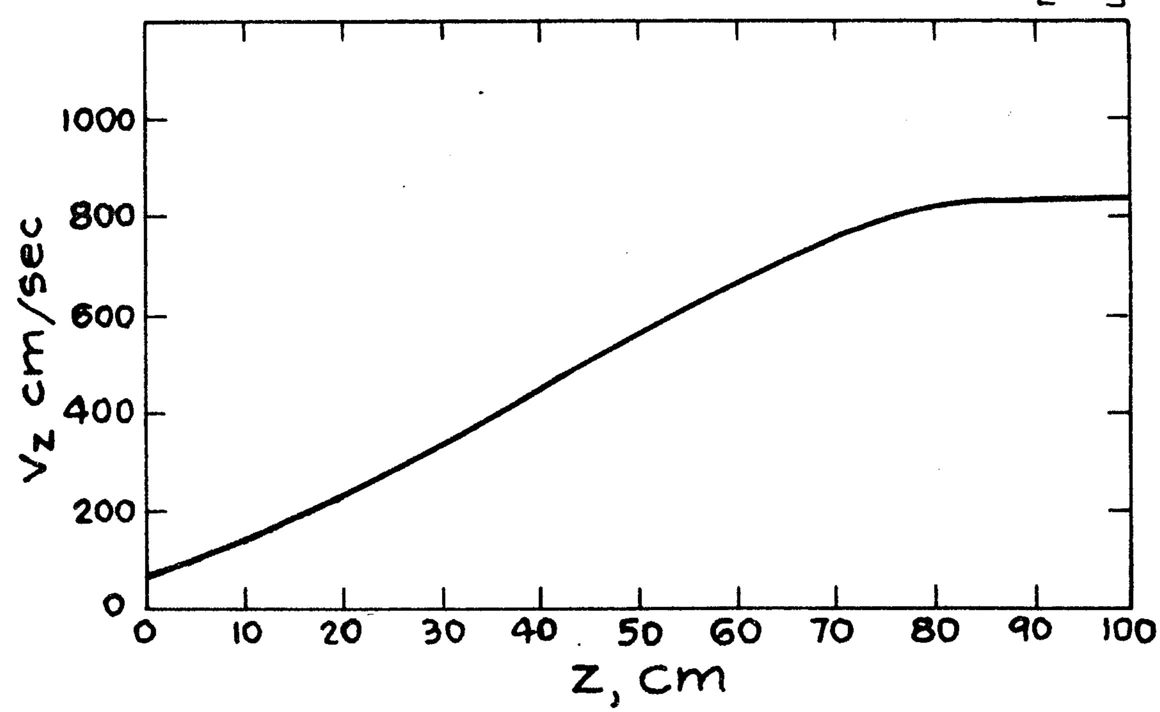

FIG. 4.15 THE VELOCITY PROFILE IN A FILAMENT OF NYLON 6.

could be a manifestation of this property? None of the non-Newtonian models examined in the present analysis predicts such results (in the absence of hardening). It is, however, possible that the use of a more general constitutive equation may lead to such results, but then, a more general constitutive equation might also make the simplified analysis of the forgoing pages no longer simple.

Although the present study has been confined to the description of the behavior of fluids in spinning processes, the principal conclusions reached here can also be applied to the formation of liquid filaments under tension in general. Thus, fluids which exhibit an elongational viscosity which increases with the rate of elongation would be capable of forming filaments under tension of lengths larger than the corresponding filaments of fluids which do not possess this property. Viscoelastic fluids which apparently possess this property are, therefore, capable of forming filaments that are appreciably longer than filaments of a Newtonian fluid of the same zero viscosity.

CHAPTER 5

THE CAPILLARY STABILITY OF A FILAMENT OF AN INCOMPRESSIBLE SIMPLE FLUID IN STEADY EXTENSION

5.1 INTRODUCTION

The linear stability analysis of chapter 4 has failed to account for the unusual stability of the thin filaments of fluid which are formed during the break-up of a visco-elastic jet. Since the filaments have been observed to elongate and thin along their flight way, we have postulated that their stable behavior might be related to the existence of a state of axial tension in the filaments. It is the purpose of this chapter, therefore, to examine the stability of a liquid filament (which is under tension) relative to small surface disturbances.

The primary flow behavior of the filaments being unknown, it is first necessary to assign, a priori, a suitable basic flow profile which is capable of describing the elongation and thinning of a cylindrical filament of fluid under tension. We have selected <u>steady extension</u> as the primary flow profile since it meets the above requirement and, in addition, proves more amenable to the subsequent stability analysis than any other likely choice for a basic profile. The rheological behavior of a filament fluid is assumed to be represented by Noll's theory of an incompressible simple fluid †. The behavior of a simple fluid in steady extension has been determined by Coleman and Noll [12] whose results are outlined in the next section.

The analysis of the stability of a filament under tension (relative to small surface disturbances) requires knowledge of the response of the fluid to small perturbations which are superimposed on the primary steady motion. It has been shown in chapter 2 that this type of response is fundamentally different than the corresponding response to small perturbations about a state of equilibrium. In section 5.3 we have, therefore, developed constitutive equations which describe the response of a simple fluid to small perturbations superposed on steady extension. The development rests upon the assumption that the matrix of physical components of the superposed strain rate consists only of diagonal terms all of which are independent of the radial direction. The resulting constitutive equations are linear in the superposed strain rate and resemble the integral-strain rate relation of infinitesimal viscoelasticity. However, in contrast to the latter, the former relations involve several memory functions which

† Henceforth, the term "simple fluid" shall imply "incompressible simple fluid".

depend on the invariants of the primary flow. With the aid of these relations, the stability of a filament of a simple fluid relative to small surface disturbances is examined in sections 5.4 and 5.5.

5.2 THE PRIMARY FLOW - STEADY EXTENSION

The steady extension of a cylindrical filament of fluid is described by the following velocity components

$$V_z = ez \;, \quad V_r = -\frac{e}{2}r \;, \quad V_\theta = 0 \tag{5.1}$$

It follows from eq. (5.1) that the filament retains its circular cylindrical shape at all times and that the length L(t) and the radius R(t) of the filament change with time according to

$$\frac{dL(t)}{dt} = e\,L(t) \tag{5.2a}$$

$$\frac{dR(t)}{dt} = -\frac{e}{2}R(t) \tag{5.2b}$$

The stresses within the filament depend on the rheological properties of the filament material. For a simple fluid Coleman and Noll show that

$$T_{rr} = -\tfrac{1}{2}\, q_1 e + \tfrac{1}{4}\, q_2 \cdot e^2 \tag{5.3}$$

$$T_{zz} = q_1 \cdot e + q_2 \cdot e^2 \tag{5.4}$$

where $q_1 = q_1(\overline{II},\overline{III})$ and $q_2 = q_2(\overline{II},\overline{III})$ are material functions of the second and third moments of the rate of strain tensor, with $\overline{II}$ and $\overline{III}$ given by

$$\overline{II} = (3/2)e^2 \quad ; \quad \overline{III} = (5/4)e^3 \tag{5.5}$$

The form of the material functions q_1 and q_2 depends on the fluid under consideration and, in general, should be determined experimentally. Thermodynamical considerations, however, place a restriction on these functions in the form,

$$(3/2)q_1 \cdot e^2 + (5/4)q_2 \cdot e^3 \geqslant 0 \tag{5.6}$$

The above inequality is in consequence of the requirement that the rate of power dissipation be non-negative.

The equations of motion (in the absence of body forces) are

r-direction

$$\rho \frac{e^2}{4} r = -\frac{\partial p}{\partial r} \tag{5.7}$$

z-direction

$$\rho e^2 z = -\frac{\partial p}{\partial z} \tag{5.8}$$

If the filament fluid is of sufficiently high viscosity, and the extension rate "e" is small, the inertia can be neglected in comparison with the contact forces. It then follows from eqs. (5.7) and (5.8) that

$$p = p(t) \tag{5.9}$$

At the free cylindrical surface the normal component of the stress is balanced by the stress induced by the surface tension force;, i.e.

$$S_{rr} = -p + T_{rr} = -\sigma/R(t) \tag{5.10}$$

where σ is the coefficient of surface tension. The effect of the ambient pressure has been neglected. Combining eqs. (5.3), (5.4) and (5.10) yields an expression for the total tensile stress, S_{zz}, in the filament.

$$S_{zz} = -p + T_{rr} = -\frac{\sigma}{R} + T_{zz} - T_{rr}$$

$$= -\frac{\sigma}{R} + \frac{3}{2}q_1 e + \frac{3}{4}q_2 e^2 \tag{5.11}$$

For a Newtonian fluid

$$q_1 = 2\eta_0 \ , \quad q_2 = 0 \tag{5.12}$$

hence

$$S_{zz} = -\frac{\sigma}{R} + 3\eta_0 e \tag{5.13}$$

For a viscoelastic fluid the material functions q_1 and q_2 must be determined experimentally. Experimental results [1, 3] indicate that the elongational viscosity defined as

$$\eta_T = S_{zz}/e \tag{5.14}$$

increases with the rate of extension "e". It should be pointed out that in the above cited experiments measurements were made on fluids of a high zero viscosity. Under these circumstances the effect of surface tension is expected to be negligible. Nonetheless, on the basis of many qualitative observations of the behavior of viscoelastic fluids under tension it is reasonable to assume that η_T would be an increasing function of the extension rate over a wider range of conditions. In any event, we would expect that the group

$$(3/2)q_1 + (3/4)q_2 \cdot e$$

be an increasing function of "e".

5.3 THE RESPONSE OF A SIMPLE FLUID TO SMALL PERTURBATIONS SUPERPOSED ON STEADY EXTENSION

We consider small perturbations superimposed on the primary steady extension such that

$$V_z = e z + V_z^{\circ}(z,t) \tag{5.15}$$

where v_z^o is the perturbed axial velocity and is assumed to be independent of the radial direction. With $v_\theta = 0$, it follows from the continuity equation that

$$V_r = -\frac{r}{2}e + V_r^{\circ} = -\frac{r}{2}e - \frac{r}{2}\frac{\partial V_z^{\circ}}{\partial z} \tag{5.16}$$

The physical components of the rate of strain tensor $\underset{\sim}{D}$ can thus be written as

$$\underset{\sim}{D} = \bar{\underset{\sim}{D}} + \underset{\sim}{D}^{o} \tag{5.17}$$

where

$$\bar{\underset{\sim}{D}} = \begin{pmatrix} -e & 0 & 0 \\ 0 & -e & 0 \\ 0 & 0 & +2e \end{pmatrix} \tag{5.18}$$

and

$$\underset{\sim}{D}^{\circ} = \begin{pmatrix} -\frac{\partial V_z^\circ}{\partial z} & 0 & -\frac{r}{2}\frac{\partial^2 V_z^\circ}{\partial z^2} \\ 0 & -\frac{\partial V_z^\circ}{\partial z} & 0 \\ -\frac{r}{2}\frac{\partial^2 V_z^\circ}{\partial z^2} & 0 & \frac{2\,\partial V_z^\circ}{\partial z} \end{pmatrix} \tag{5.19}$$

The order of magnitude of the terms appearing in eq. (5.19) can be estimated as follows

$$\frac{\partial V_z^\circ}{\partial z} = O\left(\frac{V_0}{\zeta}\right) \tag{5.20}$$

where ζ is a measure of the wave length of a typical disturbance and V_0 is a characteristic velocity which, for the present purpose, need not be specified. The off-diagonal terms in eq. (5.19) are of magnitude of the order of

$$-\frac{r}{2}\frac{\partial^2 V_z^\circ}{\partial z^2} = O\left(\frac{R}{\zeta}\cdot\frac{V_0}{\zeta}\right) \tag{5.21}$$

Now, if at all times the wave length of a typical disturbance is much larger than the radius R of the unperturbed filament, it follows from eqs. (5.20) and (5.21) that

$$\left|\frac{\partial V_z^\circ}{\partial z}\right| \gg \left|\frac{r}{2}\frac{\partial^2 V_z^\circ}{\partial z^2}\right| \tag{5.22}$$

and, hence, eq. (5.19) may be approximated by

$$\underset{\sim}{D}^{\circ} = \begin{pmatrix} -\frac{\partial v_z^{\circ}}{\partial z} & 0 & 0 \\ 0 & -\frac{\partial v_z^{\circ}}{\partial z} & 0 \\ 0 & 0 & 2\frac{\partial v_z^{\circ}}{\partial z} \end{pmatrix} \qquad (5.23)$$

The subsequent development applies to the case in which both eqs. (5.22) and (5.23) hold.

Let $\underset{\sim}{G}_t(t-s)$ be the relative strain history associated with the entire motion as described by eqs. (5.15) and (5.16), and let $\bar{\underset{\sim}{G}}_t(t-s)$ be the history corresponding to the primary steady extension. The difference between these histories

$$\overset{o}{\underset{\sim}{G}}_t(t-s) = \underset{\sim}{G}_t(t-s) - \bar{\underset{\sim}{G}}_t(t-s) \qquad (5.24)$$

corresponds, therefore, to the perturbed motion.

Turning now to the description of the stresses. Let $\bar{T}$ be the deviatoric stress of the primary motion and T^{o}, the deviatoric stress generated by the superposed purturbations. In accordance with Noll's simple fluid theory, the present value of the deviatoric stress is given by

$$\underset{\sim}{T} = \overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\underset{\sim}{G}_t(t-s)\right] \qquad (5.25)$$

where $\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ is a functional which maps tensor-valued functions into tensors. We assume that, for some suitable measure of the norm of $\overset{o}{\underset{\sim}{G}}_t(t-s)$, $\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ of eq. (5.25) can be expanded about the primary history $\bar{\underset{\sim}{G}}_t(t-s)$. The resulting expansion yields an expression for the stress in which the lowest-order term corresponds to the primary history $\bar{\underset{\sim}{G}}_t(t-s)$ and the next term is linear in the perturbed history $\underset{\sim}{G}_t^o(t-s)$. Higher-order terms have not been considered. Thus,

$$\underset{\sim}{T}(t) = \overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\bar{\underset{\sim}{G}}_t(t-s)\right] + \delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\underset{\sim}{G}_t^o(t-s); \bar{G}_t\right] \tag{5.26}$$

so that

$$\bar{\underset{\sim}{T}} = \overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\underset{\sim}{G}_t(t-s)\right] \tag{5.27}$$

and

$$\underset{\sim}{T}^o = \delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\underset{\sim}{G}_t^o(t-s); \bar{\underset{\sim}{G}}_t\right] \tag{5.28}$$

In the above expressions $\delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ are Frechet differentials [11, 14], linear in $G_t^o(t-s)$. The dependence of $\delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}$ on the primary history $\bar{\underset{\sim}{G}}_t(t-s)$ amounts to a dependence on the basic invariants of the primary motion or, equivalently, on the steady rate of extension "e". Thus

$$\underset{\sim}{T}^o = \delta\overset{\infty}{\underset{s=0}{\mathfrak{F}}}\left[\underset{\sim}{G}_t^o(t-s); e\right] \tag{5.29}$$

It can be shown [11, 14] that under suitable conditions eq. (5.29) can be represented as an integral relation in the form

$$\underset{\sim}{T}^o = \int_0^\infty M(s; e)\cdot\left\{\underset{\sim}{G}_t^o(t-s)\right\} ds \tag{5.30}$$

where $M(s;e)\cdot\{\overset{\circ}{\underset{\sim}{G}}_t(t-s)\}$ is a linear transformation of symmetric tensors into symmetric tensors. In the index notation eq. (5.30) can be expressed as

$$\overset{\circ}{T}_{ij} = \int_0^\infty M_{ijk\ell}(s;e)\,\overset{\circ}{G}_{t_{k\ell}}(t-s)\,ds \qquad (5.31)$$

We shall now obtain a simpler form of eq. (5.31) by utilizing some of the kinematic relations given in section 1.3. Recalling that $\underset{\sim}{F}(t)$ (the deformation gradient function with respect to some arbitrary reference configuration) and $\underset{\sim}{\Gamma}(t)$ (the velocity gradient tensor) are related by

$$\frac{D\underset{\sim}{F}}{Dt} = \underset{\sim}{\Gamma}\cdot\underset{\sim}{F} \qquad (5.32)$$

with, say

$$\underset{\sim}{F}(0) = \underset{\sim}{I} \qquad (5.33)$$

let $\bar{\underset{\sim}{\Gamma}}$ and $\underset{\sim}{\Gamma}^\circ$ correspond, respectively, to the components of the velocity gradient of the primary and the perturbed motions, and let $\underset{\sim}{K}^o(t)$ be defined by

$$\underset{\sim}{K}^\circ(t) = \int \underset{\sim}{\Gamma}^\circ(t)\,dt \qquad (5.34)$$

where the integration is considered indefinite. It follows from eqs. (5.32) and (5.33) that

$$\underset{\sim}{F}(t) = e^{-\underset{\sim}{K}^\circ(0)}\,e^{\bar{\underset{\sim}{\Gamma}}t + \underset{\sim}{K}^\circ(t)} \qquad (5.35)$$

In accordance with our basic assumption leading to eq. (5.23), $\mathring{\Gamma}(t)$ and $\mathbf{K}^{o}(t)$ consist both of only diagonal terms; hence,

$$\mathbf{F}^{-1}(t) = e^{+\mathbf{K}^{o}(0)} e^{-\bar{\Gamma}t - \mathbf{K}^{o}(t)} \qquad (5.36)$$

By using the relation

$$\mathbf{F}(\tau) = \mathbf{F}_t(\tau)\cdot\mathbf{F}(t) \qquad (5.37)$$

we find that

$$\mathbf{F}_t(\tau) = \mathbf{F}(\tau)\cdot\mathbf{F}^{-1}(t) = e^{-\bar{\Gamma}(t-\tau) - \mathbf{K}^{o}(t) - \mathbf{K}^{o}(\tau)} \qquad (5.38)$$

and the right relative Cauchy-Green tensor $C_t(\tau)$ is given by

$$\mathbf{C}_t(\tau) = \mathbf{F}_t^{T}(\tau)\cdot\mathbf{F}_t(\tau) = e^{-2\bar{\Gamma}(t-\tau) - 2\mathbf{K}^{o}(t) + 2\mathbf{K}^{o}(\tau)} \qquad (5.39)$$

Thus, the history of the strain $\mathbf{G}_t(\tau)$ becomes

$$\mathbf{G}_t(\tau) = \mathbf{C}_t(\tau) - \mathbf{I} = e^{-2\bar{\Gamma}(t-\tau) - 2\mathbf{K}^{o}(t) + 2\mathbf{K}^{o}(\tau)} - \mathbf{I} \qquad (5.40)$$

or, by letting

$$\tau = t - s \qquad (5.41)$$

$\mathbf{G}_t(\tau)$ becomes

$$\mathbf{G}_t(t-s) = e^{-2\bar{\Gamma}s - 2\mathbf{K}^{o}(t) + 2\mathbf{K}^{o}(t-s)} - \mathbf{I} \qquad (5.42)$$

For small $\mathbf{K}^{o}(\;)$, eq. (5.42) can be approximated by

$$\mathbf{G}_t(t-s) = e^{-\bar{\Gamma}s}\,[\mathbf{I} - 2\mathbf{K}^{o}(t)][\mathbf{I} + 2\mathbf{K}^{o}(t-s)] - \mathbf{I} \qquad (5.43a)$$

or

$$\underset{\sim}{G}_t(t-s) = e^{-2\bar{\underset{\sim}{\Gamma}} s} [\underset{\sim}{I} - 2\underset{\sim}{K}^{\circ}(t) + 2\underset{\sim}{K}^{\circ}(t-s)] - \underset{\sim}{I} \qquad (5.43b)$$

In the above, terms of order $\|\underset{\sim}{K}^{o}(\ )\|$, where $\|\underset{\sim}{K}^{o}(\ )\|$ is some suitable norm of $\underset{\sim}{K}^{o}(\ )$, have been neglected. From eq. (5.42) it is clear that the primary history $\bar{\underset{\sim}{G}}_t(t-s)$ (corresponding to the case in which $\underset{\sim}{K}^{o}(\ ) = \underset{\sim}{0}$) is

$$\bar{\underset{\sim}{G}}_t(t-s) = e^{-2\bar{\underset{\sim}{\Gamma}} s} \qquad (5.44)$$

The difference history $\overset{o}{\underset{\sim}{G}}_t(t-s)$ is therefore given by

$$\overset{\circ}{\underset{\sim}{G}}_t(t-s) = \underset{\sim}{G}_t(t-s) - \bar{\underset{\sim}{G}}_t(t-s) = 2e^{-2\bar{\underset{\sim}{\Gamma}} s}[\underset{\sim}{K}^{\circ}(t-s) - \underset{\sim}{K}^{\circ}(t)] \qquad (5.45)$$

Eq. (5.45) is now substituted in the constitutive equation of (5.30) giving

$$\underset{\sim}{T}^{\circ}(t) = 2\int_0^{\infty} M(s;e)\cdot\left\{e^{-2\bar{\underset{\sim}{\Gamma}} s}[\underset{\sim}{K}^{\circ}(t-s) - \underset{\sim}{K}^{\circ}(t)]\right\} ds$$

$$= 2\Phi(0,e)\left\{\underset{\sim}{K}^{\circ}(t)\right\} + 2\int_0^{\infty} M(s;e)\left\{e^{-\bar{\underset{\sim}{\Gamma}} s}\underset{\sim}{K}^{\circ}(t-s)\right\} ds \qquad (5.46)$$

where

$$\frac{d\Phi}{ds}(s,e) = -M(s;e)\left\{e^{-\bar{\underset{\sim}{\Gamma}} s}\right\} \qquad (5.47)$$

Integrating eq. (5.48) by parts yields

$$\underset{\sim}{T}^{\circ}(t) = 2\int_0^{\infty} \Phi(s,e)\left\{\underset{\sim}{\Gamma}^{\circ}(t-s)\right\} ds \qquad (5.48)$$

or

$$\underset{\sim}{T}^{\circ}(t) = \int_0^{\infty} \Phi(s,e)\left\{ \underset{\sim}{D}^{\circ}(t-s)\right\} ds \qquad (5.49)$$

Eq. (5.49) will henceforth serve as the constitutive equation describing small perturbations superposed on steady extension. It is convenient at this point to get expressions for two of the components of the perturbed stress. Thus, using eq. (5.23),

$$\overset{\circ}{T}_{rr} = \overset{\circ}{T}_{11} = \int_0^{\infty}\left(- \Phi_{1111}\frac{\partial \overset{\circ}{V}_z}{\partial z} - \Phi_{1122}\frac{\partial \overset{\circ}{V}_z}{\partial z} + 2\Phi_{1133}\frac{\partial \overset{\circ}{V}_z}{\partial z}\right) ds$$

$$= \int_0^{\infty} - \Psi_1(s,e)\frac{\partial \overset{\circ}{V}_z}{\partial z}(t-s)\, ds \qquad (5.50)$$

where

$$\Psi_1 = \Phi_{1111} + \Phi_{1122} - 2\Phi_{1133} \qquad (5.51)$$

Likewise

$$\overset{\circ}{T}_{zz} = \overset{\circ}{T}_{33} = \int_0^{\infty}\left(- \Phi_{3311}\frac{\partial \overset{\circ}{V}_z}{\partial z} - \Phi_{3322}\frac{\partial \overset{\circ}{V}_z}{\partial z} + \Phi_{3333}\frac{\partial \overset{\circ}{V}_z}{\partial z}\right) ds$$

$$= \int_0^{\infty} 2\Psi_3(s,e)\frac{\partial \overset{\circ}{V}_z}{\partial z}(t-s)\, ds \qquad (5.53)$$

where

$$2\Psi_3 = - \Phi_{3311} - \Phi_{3322} + 2\Phi_{3333} \qquad (5.54)$$

Further, let

$$T^{\circ}_{zz} - T^{\circ}_{rr} = \int_0^{\infty} [2\Psi_3(s,e) - \Psi_1(s,e)] \frac{\partial V^{\circ}_z}{\partial z}(t-s)\,ds$$

$$= \int_0^{\infty} \Psi(s,e) \frac{\partial V^{\circ}_z}{\partial z}(t-s)\,ds \qquad (5.55)$$

The various components of Φ are, of course, not all independent. Thus, symmetry implies that

$$\Phi_{ijk\ell} = \Phi_{jik\ell} \qquad (5.56)$$

and

$$\Phi_{ijk\ell} = \Phi_{ij\ell k} \qquad (5.57)$$

The fact that $\underset{\sim}{T}^{o}$ is deviatoric also requires that

$$\sum_i \Phi_{iik\ell} = 0 \qquad (5.58)$$

Other consistency relations between the components of Φ may still be obtained. However, for the present purposes, eqs. (5.50), (5.53) and (5.55) in their present form are deemed sufficient.

Note that for a Newtonian fluid of viscosity η_o

$$T^{\circ}_{rr} = -\eta_o \frac{\partial V^{\circ}_z}{\partial z} \qquad (5.59a)$$

and

$$T^{\circ}_{zz} = 2\eta_o \frac{\partial V^{\circ}_z}{\partial z} \qquad (5.59b)$$

so that

$$T_{zz}^{\circ} - T_{rr}^{\circ} = 3\eta_0 \frac{\partial V_z^{\circ}}{\partial z} \tag{5.59c}$$

Also, when e = 0, eq. (5.55) reduces to,

$$T_{zz}^{\circ} - T_{rr}^{\circ} = \int 3\,\varphi(s) \frac{\partial V_z^{\circ}}{\partial z}(t-s)\,ds \tag{5.60}$$

where $\varphi(s)$ is the relaxation function of infinitesimal viscoelasticity whose Laplace transform is

$$\hat{\varphi} = \hat{\eta}(\alpha) \tag{5.61}$$

where $\hat{\eta}(\alpha)$ is the complex viscosity of eq. (2.3).

5.4 SOLUTION OF THE DYNAMIC EQUATIONS

We summarize below the basic assumptions which have guided the development of the previous sections:

a) The inertia of the primary flow is negligible.

b) $\partial V_z^{\circ}/\partial r = 0$

c) At all times the wave length of a typical disturbance is much larger than the unperturbed radius of the filament; this implies that $V_z^{\circ} >> V_r^{\circ}$.

d) The disturbances are axisymmetric.

e) There are no interactions with the surrounding medium.

In view of these assumptions, we could employ here the momentum balance of the previous chapter (dealing with spinning) which have been developed under similar circumstances.

Thus, using eq. (4.27), the momentum equation becomes,

$$\rho V_z \frac{\partial V_z}{\partial z} \frac{(R+\xi)^2}{2} = \frac{\partial}{\partial z}\left[(-p + T_{zz})\frac{(R+\xi)^2}{2}\right] - \rho \frac{\partial V_z}{\partial t}\frac{(R+\xi)^2}{2}$$

$$+ (R+\xi)\left[T_{rz} - (-p + T_{zz})\frac{\partial \xi}{\partial z}\right]_{r=R+\xi} \qquad (5.62)$$

where $\xi = \xi(z,t)$ is a small disturbance superposed on the surface of the filament. It is assumed that at all times

$$\xi << R \qquad (5.63)$$

The boundary conditions at the free surface of the filament are

$$\left[T_{rz} - (-p + T_{zz})\frac{\partial \xi}{\partial z}\right]_{r=R+\xi} = -2H\sigma \frac{\partial \xi}{\partial z} \qquad (5.64)$$

$$\left[-p + T_{rr}\right]_{r=R+\xi} = 2H\sigma \qquad (5.65)$$

where

$$2H\sigma = -\frac{\sigma}{R+\xi} + \sigma \frac{\partial^2 \xi}{\partial z^2} \qquad (5.66)$$

or, by expanding the first term on the right of eq. (5.66) in terms of ξ/R and neglecting second-order in ξ/R,

$$2H\sigma = -\frac{\sigma}{R} + \frac{\sigma}{R^2}\left(\xi + R^2 \frac{\partial^2 \xi}{\partial z^2}\right) \qquad (5.67)$$

In addition, the following dynamic equation holds at the free surface of the jet:

$$\frac{D(R+\xi)}{Dt} = V_r \Big|_{r=R+\xi} \qquad (5.68)$$

or

$$\frac{dR}{dt} + \frac{\partial \xi}{\partial t} + V_z \frac{\partial \xi}{\partial z} = -\frac{(R+\xi)}{2}\frac{\partial V_z}{\partial z} \tag{5.69}$$

Substituting eqs. (5.64) and (5.65) into the equation of motion of (5.62) yields

$$\rho V_z \frac{\partial V_z}{\partial z}\frac{(R+\xi)^2}{2} + \rho \frac{\partial V_z}{\partial t}\frac{(R+\xi)^2}{2} = \frac{\partial}{\partial z}\left[(T_{zz} - T_{rr} + 2H\sigma)\frac{(R+\xi)^2}{2}\right] - 2H\sigma(R+\xi)\frac{\partial \xi}{\partial z} \tag{5.70}$$

Substituting into eq. (5.70) the following expressions

$$V_z = ez + V_z^\circ \tag{5.71a}$$

$$\frac{\partial V_z}{\partial z} = e + \frac{\partial V_z^\circ}{\partial z} \tag{5.71b}$$

$$T_{zz} = \overline{T}_{zz} + T_{zz}^\circ \tag{5.71c}$$

$$T_{rr} = \overline{T}_{rr} + T_{rr}^\circ \tag{5.71d}$$

and equating first-order terms in the perturbed quantities gives the following expression for the equation of motion of the perturbed flow:

$$\rho \frac{R^2}{2}\left(\frac{\partial V_z^\circ}{\partial t} + ez\frac{\partial V_z^\circ}{\partial z} + eV_z^\circ\right) = R(\overline{T}_{zz} - \overline{T}_{rr})\frac{\partial \xi}{\partial z} + \frac{R^2}{2}\frac{\partial}{\partial z}(T_{zz}^\circ - T_{rr}^\circ) + \frac{\sigma}{2}\left(\frac{\partial \xi}{\partial z} + R^2\frac{\partial^3 \xi}{\partial z^3}\right) \tag{5.72}$$

Eq. (5.69) similarly yields

$$\frac{\partial \xi}{\partial t} + ez\frac{\partial \xi}{\partial z} = -\frac{R}{2}\frac{\partial V_z^\circ}{\partial z} - \frac{e}{2}\xi \tag{5.73}$$

For convenience, let

$$T \equiv T_{zz} - T_{rr} \qquad (5.74a)$$

so that

$$\bar{T} = \bar{T}_{zz} - \bar{T}_{rr} \qquad (5.74b)$$

and

$$T^{\circ} = T^{\circ}_{zz} - T^{\circ}_{rr} \qquad (5.74c)$$

Eq. (5.72) thus becomes

$$\rho \frac{R^2}{2}\left(\frac{\partial V_z^{\circ}}{\partial t} + e z \frac{\partial V_z^{\circ}}{\partial z} + e V_z^{\circ}\right) = R\bar{T}\frac{\partial \xi}{\partial z} + \frac{R^2}{2}\frac{\partial T^{\circ}}{\partial z} + \frac{\sigma}{2}\left(\frac{\partial \xi}{\partial z} + R^2 \frac{\partial^3 \xi}{\partial z^3}\right) \qquad (5.75)$$

In the above equation

$$\bar{T} = \frac{3}{2} q_1 e + \frac{3}{4} q_2 e^2 \qquad (5.76)$$

and T^{o} is given by eq. (5.55); i.e.

$$T^{\circ} = \int_0^{\infty} \Psi(s,e) \frac{\partial V_z^{\circ}}{\partial z}(t-s)\, ds \equiv \left[\Psi * \frac{\partial V_z^{\circ}}{\partial z}\right] \qquad (5.77)$$

Eq. (5.73), written in the form

$$\frac{\partial V_z^{\circ}}{\partial z} = -\frac{2}{R}\left(\frac{\partial \xi}{\partial t} + e z \frac{\partial \xi}{\partial z} + \frac{e}{2}\xi\right) \qquad (5.78)$$

can be used to eliminate from eq. (5.75) terms which involve v_z^o and $\partial v_z^o/\partial z$. For this purpose, eq. (5.75) is first differentiated with respect to z. Substituting eq. (5.78) into the resulting equation yields (after a considerable manipulation) a partial differential equation involving the surface perturbation ξ only:

$$\frac{\partial^2 \xi}{\partial t} + 3e\frac{\partial \xi}{\partial t} + 2ez\frac{\partial^2 \xi}{\partial z \partial t} + 4e^2 z\frac{\partial \xi}{\partial z} + e^2 z^2 \frac{\partial^2 \xi}{\partial z^2} + \frac{5}{4}e^2 \xi = \bar{T}\frac{\partial^2 \xi}{\partial z^2} +$$

$$+\frac{1}{\rho}\left[\Psi^*\left(\frac{\partial^3 \xi}{\partial t \partial z^2} + ez\frac{\partial^3 \xi}{\partial z^3} + \frac{5}{2}e\frac{\partial^2 \xi}{\partial z^2}\right)\right] + \frac{\sigma}{2\rho R}\left(\frac{\partial^2 \xi}{\partial z^2} + R^2\frac{\partial^4 \xi}{\partial z^4}\right) \quad (5.79)$$

Eq. (5.79) is a linear partial differential equation with coefficients which depend on z and t. A full solution to this equation can not be obtained at present. However, a reasonable approximation can be readily obtained by noting that the only coefficients which depend on time appear in the last term on the right of eq. (5.79); i.e.

$$\frac{\sigma}{2\rho R}\left(\frac{\partial^2 \xi}{\partial z^2} + R^2\frac{\partial^4 \xi}{\partial z^4}\right)$$

where $R = R(t) = \exp.(-et/2)$. This term represents the action of the capillary force which indeed depends, at any moment, on the radius R of the filament. However, for small values of "e", an approximate solution can be obtained by treating R as a running parameter and getting solutions to eq. (5.79) for different values of R.

With R considered as a constant, the stability question can now be completely determined by obtaining the solution to equation (5.79) which holds in the vicinity of $z = 0$. It will subsequently be shown that the characteristic equation thus obtained is not restricted to $z = 0$, but holds identically at any position $z \neq 0$ along the filament. Accordingly, in the vicinity of $z = 0$ the solution to eq. (5.79) is of the form

$$\xi = \xi_o e^{\alpha t + ikz} \tag{5.80}$$

where α and k can be interpreted, respectively, as the growth-rate and the wave number of a disturbance-wave. Substituting eq. (5.80) into eq. (5.79) (with $z = 0$) yields

$$\alpha^2 + \alpha\left[\frac{\hat{\eta}(\alpha,e)k^2}{\rho} + 3e\right] = \frac{Tk^2}{\rho} + \frac{\sigma k^2}{2\rho R}(1 - k^2R^2) - \frac{5}{2}\frac{\hat{\eta}(\alpha,e)k^2}{\rho}e - \frac{5}{4}e^2 \tag{5.81}$$

where $\hat{\eta}(\alpha,e)$, given by

$$\hat{\eta}(\alpha,e) = \int_0^\infty \Psi(s,e)e^{-\alpha s}\,ds \tag{5.82}$$

is the complex viscosity of the complex motion.

Before proceeding to evaluate eq. (5.81), let us show that eq. (5.81) indeed represents the characteristic equation at any point ($z \neq 0$) along the filament. For this purpose, the following change of variables is introduced:

$$x = z - L(t) \tag{5.83}$$

$$t' = t \tag{5.84}$$

where L(t) is the distance of a particular material point from the origin. The change of variables is illustrated in the following diagram in which the filament is shown at two successive times:

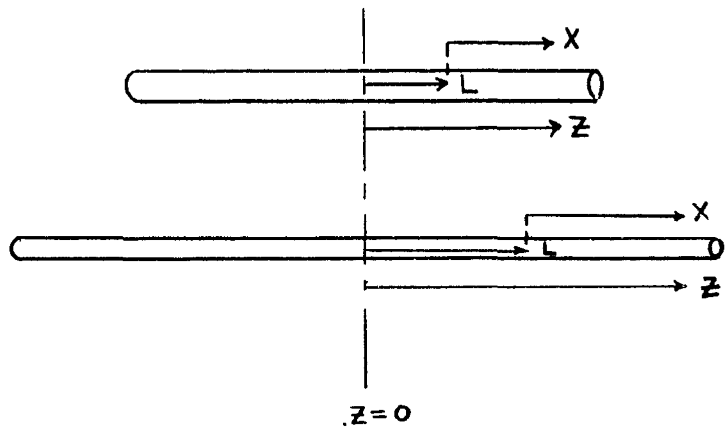

Fig. 5.1 Change of variables.

In consequence of the change of variables

$$\frac{\partial V_z^0}{\partial t} = \frac{\partial V_z^0}{\partial t'} - \frac{dL}{dt'}\frac{\partial V_z^0}{\partial x} \tag{5.85}$$

$$\frac{\partial V_z^0}{\partial z} = \frac{\partial V_z^0}{\partial x} \tag{5.86}$$

Eqs. (5.85) and (5.86) are substituted into eqs. (5.75) and (5.78) giving

$$\rho\frac{R^2}{2}\left(\frac{\partial V_z^0}{\partial t'} - \frac{dL}{dt'}\frac{\partial V_z^0}{\partial x} + eL\frac{\partial V_z^0}{\partial x} + ex\frac{\partial V_z^0}{\partial x} + eV_z^0\right) = R\bar{T}\frac{\partial \xi}{\partial x} + $$

$$+ \frac{R^2}{2}\frac{\partial T^0}{\partial x} + \frac{\sigma}{2}\left(\frac{\partial \xi}{\partial x} + R^2\frac{\partial^3 \xi}{\partial x^3}\right) \tag{5.87}$$

$$\frac{\partial V_z^0}{\partial x} = -\frac{2}{R}\left(\frac{\partial \xi}{\partial t'} - \frac{dL}{dt'}\frac{\partial \xi}{\partial x} + eL\frac{\partial \xi}{\partial x} + ex\frac{\partial \xi}{\partial x} + \frac{e}{2}\xi\right) \tag{5.88}$$

However, it follows from eq. (5.2a) that

$$\frac{dL}{dt'} = eL$$

hence, eqs. (5.87) and (5.88) reduce to

$$\frac{\rho R^2}{2}\left(\frac{\partial V_z^\circ}{\partial t'} + e x \frac{\partial V_z^\circ}{\partial x} + e V_z^\circ\right) = R\bar{T}\frac{\partial \xi}{\partial x} + \frac{R^2}{2}\frac{\partial T^\circ}{\partial x} + \frac{\sigma}{2}\left(\frac{\partial \xi}{\partial x} + R^2\frac{\partial^3 \xi}{\partial x^3}\right) \tag{5.89}$$

$$\frac{\partial V_z^\circ}{\partial x} = -\frac{2}{R}\left(\frac{\partial \xi}{\partial t'} + e x \frac{\partial \xi}{\partial x} + \frac{e}{2}\xi\right) \tag{5.90}$$

Eqs. (5.89) and (5.90) are obviously identical in form to the corresponding equations in the original variables z and t; i.e. eqs. (5.75) and (5.78). Therefore, if we proceed along the same steps which have been taken before, we would obtain at x = 0 (and, hence, at z = L) a characteristic equation which is identical to (5.81). Since L(t) has been chosen arbitrarily, the characteristic equation of (5.81) must, therefore, hold at any point along the filament. That is to say, the growth of an unstable disturbance and its corresponding wave length are unaffected by L(t) and are identical in value anywhere along the filament.

Note that the equivalent of eq. (5.80) in the new variables is

$$\xi = \xi_\circ e^{\alpha t + ikx} \tag{5.91}$$

or

$$\xi = \xi_\circ e^{\alpha t - ikL(t) + ikz} \tag{5.92}$$

From eq. (5.92) it is clear that the dependence on L(t) is manifested only as an arbitrary phase angle which is irrelevant to the stability question.

5.5 THE CHARACTERISTIC EQUATION

For convenience we rewrite the characteristic equation

$$\alpha^2 + \alpha\left[\frac{\hat{\eta}(\alpha,e)k^2}{\rho} + 3e\right] = \frac{\bar{T}k^2}{\rho} + \frac{\sigma k^2}{2\rho R}(1-k^2R^2) - \frac{5}{2}\frac{\hat{\eta}(\alpha,e)}{\rho}k^2e - \frac{5}{4}e \qquad (5.81)$$

Note first that when e = 0, eq. (5.81) reduces to the characteristic equation which has been previously derived for a cylindrical filament at rest (3.22); i.e.

$$\alpha^2 + \frac{3\hat{\eta}(\alpha)k^2\alpha}{\rho} = \frac{\sigma k^2}{2\rho R}(1-k^2R^2) \qquad (5.93)$$

To facilitate a comparison between eqs. (5.81) and (5.93), we cast them in the following form:

$$\alpha^2 = \Sigma - \frac{k^2}{\rho}\left[\frac{5}{2}\hat{\eta}(\alpha,e)e - \bar{T}\right] - \frac{5}{4}e^2 - \alpha\left[\frac{\hat{\eta}(\alpha,e)k^2}{\rho} + 3e\right] \qquad (5.94)$$

$$\alpha^2 = \Sigma \qquad\qquad\qquad\qquad - \alpha\frac{3\hat{\eta}(\alpha)}{\rho}k^2 \qquad (5.95)$$

where Σ denotes the quantity

$$\Sigma = \frac{\sigma k^2}{2\rho R}(1-k^2R^2) \qquad (5.96)$$

Note that $\Sigma > 0$ since it has been assumed that $kR \ll 1$.

Consider first the case of a Newtonian fluid for which

$$\hat{\eta}(\alpha,e) = 3\hat{\eta}(\alpha) = 3\eta_0 \qquad (5.97)$$

and

$$\bar{T} = 3\eta_0 e \qquad (5.98)$$

Eqs. (5.94) and (5.95) therefore become

$$\alpha^2 = \Sigma - \frac{4.5}{\rho}\eta_0 k^2 e - \alpha\left(\frac{3}{\rho}\eta_0 k^2 + 3e\right) \qquad (5.99)$$

$$\alpha^2 = \Sigma - \alpha\frac{3}{\rho}\eta_0 k^2 \qquad (5.100)$$

(the term $\frac{5}{4}e^2$, being always of secondary magnitude, has been omitted). In fig. 5.2 both sides of eqs. (5.99) and (5.100) have been plotted as functions of α (for fixed values of k and R). In this figure, the solid straight line represents the right-hand side of eq. (5.99) (a filament under tension, $e \neq 0$), while the dotted line corresponds to the right-hand side of eq. (5.100) (a filament at rest, e = 0). From this figure it is clear that the magnitude of α corresponding to a filament under tension (point 1 in fig. 5.2) is <u>always smaller</u> than the corresponding eigen value α for a filament at rest (point 2 in fig. 5.2). Furthermore, whereas a filament of a Newtonian fluid at rest in unconditionally unstable, a filament of a Newtonian fluid undergoing steady extension may be completely stable as long as

$$\frac{4.5}{\rho}\eta_0 k^2 e \geqslant \Sigma \qquad (5.101)$$

or, equivalently

$$9\eta_0 e \geqslant \frac{\sigma}{R} \qquad (5.102)$$

However, since a filament in steady extension thins continually, the value of R decreases and the inequality of (5.102)

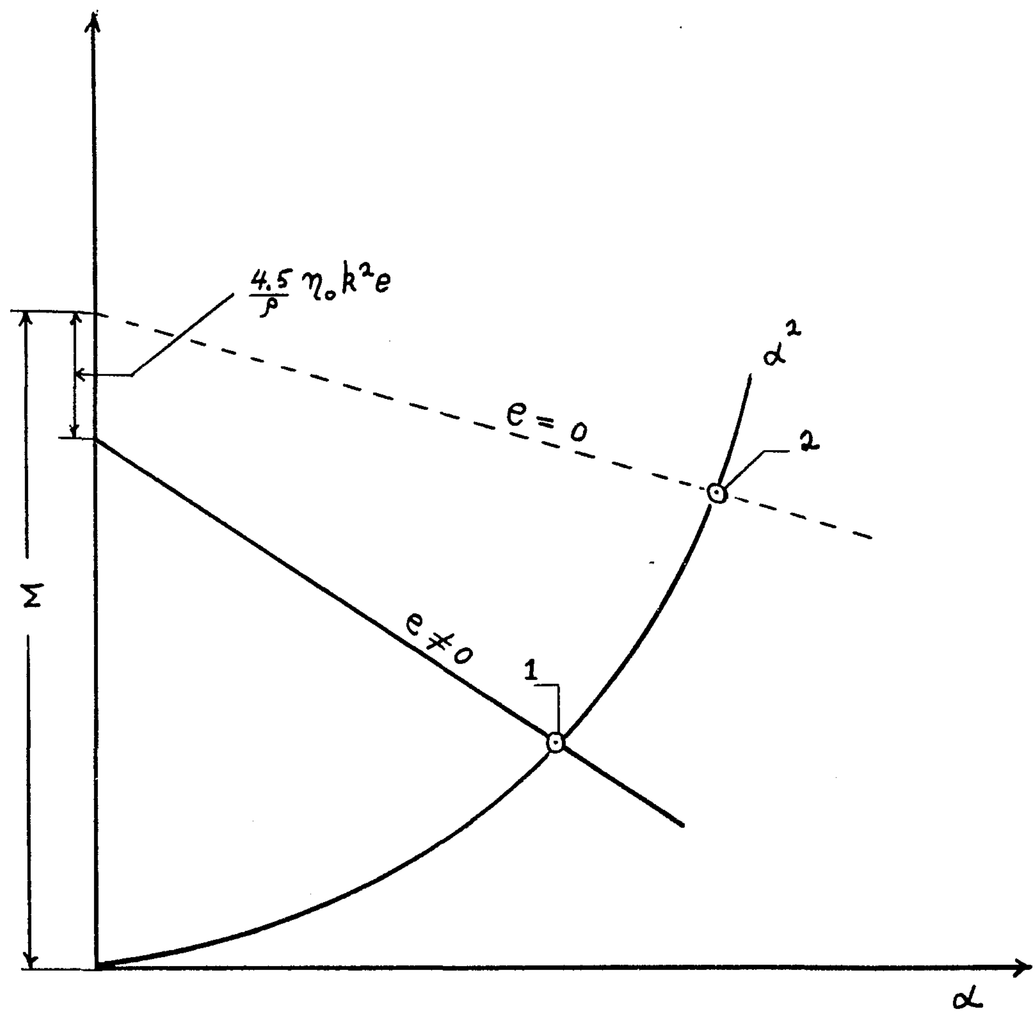

Fig. 5.2 Qualitative representation of eqs. (5.99) and (5.100).

will eventually be reversed. The filament will then become unstable and subsequently break-up into droplets. The relative magnitudes of α for a filament of a Newtonian fluid in steady extension and at rest are also compared in fig. 5.3 in which a portion of the plot of α vs. k^2 is shown for a fixed value of R. The figure indeed shows that as the value of "e" increases, the entire spectrum of eigen values α decrease in magnitude.

Thus, on the basis of the above results, one would expect that the stability of a Newtonian filament (relative to small surface disturbances) would be enhanced when the filament is subjected to axial tension. Is there any physical evidence which could corroborate this conclusion? Admittedly, conclusive evidence does not exist; however, two qualitative observations might be cited in support of the above results. The first observation was made by Donnelly and Glaberson [20] who noted that the break-up length of a Newtonian jet which is ejected vertically downwards is considerably greater than the corresponding break-up length for a horizontal jet. The higher stability in this case might be due to the axial tension to which the jet is subjected in consequence of the action of gravity. Perhaps a more convincing argument is offered in fig. 5.4. Fig. 5.4 is a photograph of a thin filament (of the order of 10^{-3} cm in diameter) of a Newtonian

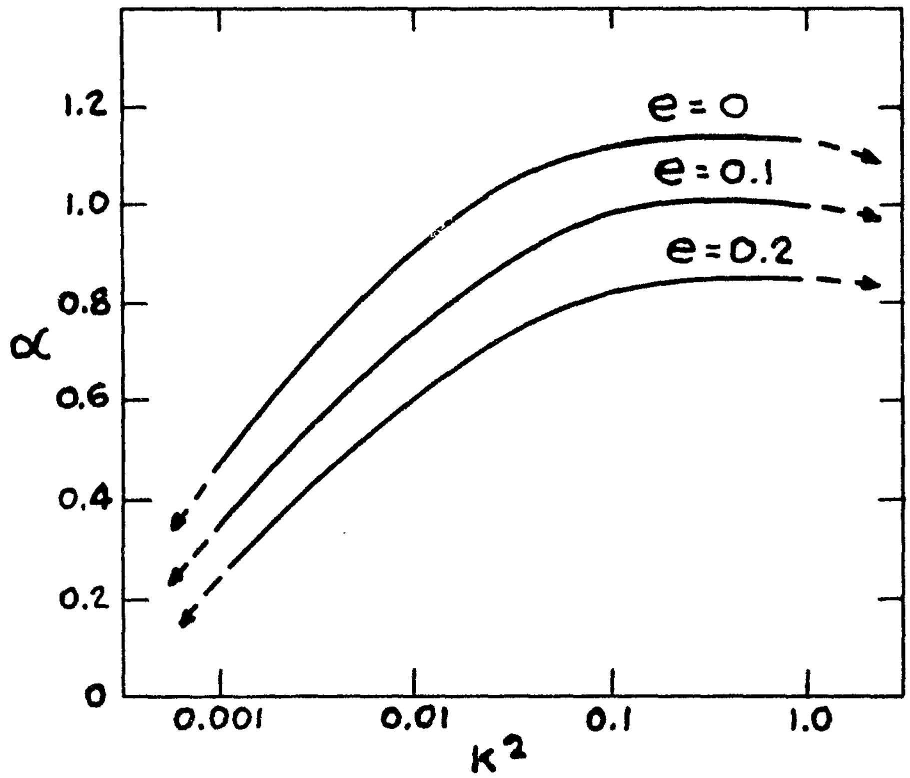

FIG. 5.3 α VS K^2 FOR A FILAMENT OF A NEWTONIAN FLUID AT REST ($e = 0$), AND IN STEADY EXTENSION ($e = 0.1$, $e = 0.2$); $\eta_0 = 100$ poise, $\rho = 1$ gm/cm^3, $R = 0.1$cm, $\sigma = 70$ dyne/cm

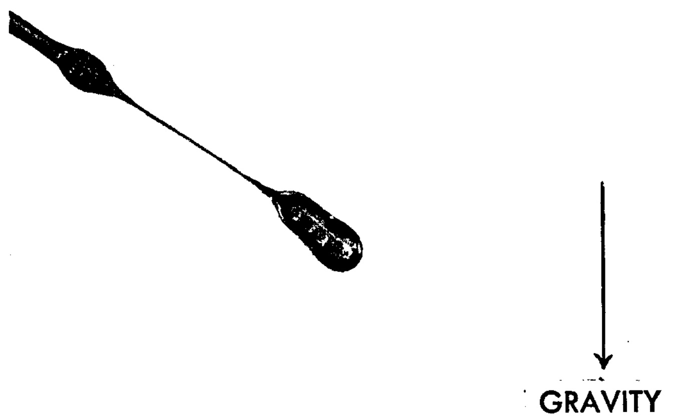

Fig. 5.4 A Filament of a Newtonian Fluid under tension (Original Jet Diameter — 0.0216 cm).

fluid (glycerine) which has formed during the break-up of a jet. The filament, as shown, is undoubtedly under tension caused by the weight of the lower droplet to which the filament is attached. Comparable threads of fluid have not been observed during the break-up of horizontal Newtonian jets. Further, the filament is considerably longer than the length predicted by the theoretical results for a filament at rest.

Turning now to the case of a viscoelastic fluid. It is clear that the stability question depends on the relative magnitudes of $\bar{T}$ and $\hat{\eta}(\alpha,e)e$. Since both $\bar{T}$ and $\hat{\eta}(\alpha,e)$ are expressed in terms of unknown material functions, an explicit solution to the characteristic equation can not be obtained. Further, since no information is available on the nature of the complex viscosity $\hat{\eta}(\alpha,e)$, evaluation of the characteristic equation can be made only by means of general conjectures which rest, insofar as it is possible, on physical experience. Thus, physical evidence indicates that

$$\bar{T} = (3/2)q_1 \cdot e + (3/4)q_2 \cdot e^2$$

is an increasing function of the extension rate "e". Therefore,

$$\bar{T} > 3\eta_o e \qquad (5.103)$$

where η_o is the zero viscosity of the fluid; i.e.

$$\lim_{e \to o} \left(\frac{\bar{T}}{e}\right) = 3\eta_o \qquad (5.104)$$

It has also been shown in chapter 2 that the complex viscosity which represents the response of viscoelastic fluids to small perturbations superimposed on steady shear flow is drastically different than the complex viscosity which describes perturbations near equilibrium. In particular (in reference to fig. 2.3), it has been noted that while the absolute value of the latter decreases monotonically with the frequency, the absolute value of the complex viscosity of the superposed oscillatory motion can exhibit a maximum. Further, the experimental results cited in section 2.3 show that the curve of the absolute value of the complex viscosity remains virtually "flat" over a wide range of frequencies before "curving down" at the long end of the frequency range. Now, there is no a priori reason, of course, to expect that the complex viscosity $\hat{\eta}(\alpha,e)$ which describes perturbations on steady extension would exhibit a similar behavior. However, in the absence of any evidence to the contrary, it is reasonable to assume that $\hat{\eta}(\alpha,e)e$ would be of comparable magnitude to $\bar{T}$ over some finite range of α. Suppose, therefore, that we envision a situation in which the filament instability is dominated by a latent root α which falls within the range where

$$\bar{T} \sim \hat{\eta}(\alpha,e)e \tag{5.105}$$

In this case the characteristic equation can be approximated by

$$\alpha^2 = \Sigma - \frac{3}{2}\frac{\bar{T}k^2e}{\rho} - \alpha\left(\frac{\bar{T}k^2}{\rho e} + 3e\right) \tag{5.106}$$

Now, it follows from eq. (5.105) that

$$\bar{T} > 3\eta_0$$

therefore, in comparison to a filament of a Newtonian fluid of viscosity η_0, the viscoelastic filament would exhibit a higher stability. This being merely a plausible conjecture, it would be futile to elaborate on these results any further without the benefit of any experimental data pertaining to $\hat{\eta}(\alpha, e)$. Nevertheless, the arguments presented above lend a substantial support to our original assertion, namely, that the observed stability of liquid filaments which are formed during the break-up of viscoelastic jets is due to a state of axial tension which exists along the filaments.

CHAPTER 6

SUMMARY AND CONCLUSIONS

The behavior of a filament of a viscoelastic fluid under tension and at rest has been studied. Three major objectives have been pursued:

(1) The stability of a filament at rest with respect to small surface disturbances.

(2) The formation of liquid filaments under steady state conditions.

(3) The stability of a filament under tension relative to small surface disturbances.

The fulfillment of these objectives depends, a priori, on the proper description of the rheological properties of the filament fluid. Accordingly, a considerable attention has been devoted to this aspect of the work with special emphasis placed upon the description of small perturbations superimposed on a fluid which is initially at rest or undergoing some primary steady motion.

The stability of a filament which is initially at rest has been studied in reference to the phenomenon of the break-up of capillary jets. The analysis closely parallels Weber's corresponding development for Newtonian jets which, indeed, constitutes a special case of the present theory.

The response of the jet to small perturbations has been represented by the complex viscosity $\hat{\eta}(\alpha)$ derived from Biot's theory of infinitesimal viscoelasticity [5]. This description is based on thermodynamic principles and is considered a valid representation of any viscoelastic material in the vicinity of equilibrium.

The results of the linear theory show that a jet of a viscoelastic fluid which is characterized by a complex viscosity that is bounded in absolute value, is less stable than a jet of a Newtonian fluid of the same zero viscosity. Experiments in weakly viscoelastic fluids confirm this result. However, data on fluids with more pronounced elastic properties indicate that non-linear phenomena are dominating. The disturbances appear as a series of large droplets connected by random lengths of thin filaments of fluid which thin continually with the distance eventually leading to the break-up of the jet. The formation and the unusual stability of these filaments is not accounted for by the theoretical results. Even in dilute viscoelastic solutions, the break-up of the jet does not occur by the growth of clearly defined waves, as is the case for jets of Newtonian fluids.

The steady state behavior of a filament under tension has been studied within the context of fiber spinning processes. The principal aim of this study has been to examine the spinnability of different fluids in relation to their inherent rheological character. To this end, energy and mass transfer between the filament and its surrounding (which dominate an actual spinning process) has been omitted. The problem has been further reduced to the case in which the mechanics of the flow are predominantly governed by the balance of axial forces. The resulting solutions are, therefore, valid for situations in which the variation of the filament radius along the spinning way is small.

The results of this study show that a filament of a Newtonian fluid of a size comparable to that of a typical synthetic fiber (produced in most commercial spinning processes) can be formed without hardening only if the viscosity of the fluid is extremely high (10^5 - 10^6 poise). Newtonian fluids in this viscosity range can not, however, be extruded through the narrow spinnerette holes without the application of an enormous pressure difference. One is therefore led to the conclusion that, without hardening, the spinning of a filament of a Newtonian fluid is not feasible.

In the case of viscoelastic fluids, the analysis indicates that the dominant property which account for their (physically observed) superior spinnability is the increase of the elongational viscosity with the elongation rate.

Fluids which exhibit a strong increase in the elongational viscosity with the rate of elongation should, hence, be capable of producing filaments without hardening. Because of the relative complexity of the present analysis, the non-Newtonian fluids which have been examined have been represented by simple fluid models. Of these, only the Reiner-Rivlin fluid exhibits an elongational viscosity which increases with the elongation rate. The two Oldroyd fluids which have been examined exhibit the opposite effect and, hence, appear less spinnable than a Newtonian fluid of the same zero viscosity. Since this is contrary to the physical evidence, the constitutive equations of the Oldroyd fluids (eqs. 4.63 and 4.64) should not be regarded as proper representatives of viscoelastic fluids in spinning processes. A more realistic description of the spinning of viscoelastic fluids would, therefore, require the testing of other constitutive equations which, in addition to their conformity to the viscoelastic phenomena which have already been understood, are also capable of exhibiting an apparent viscosity which increases with the axial deformation rate. Finally, although the present study has been confined to the description of the behavior of fluids in spinning processes, the principal conclusions reached here can also be applied to the formation of liquid filaments under tension in general.

Thus, fluids which exhibit an elongational viscosity which increases with the rate of elongation would be capable of forming filaments under tension of lengths larger than the corresponding filaments of fluids which do not possess this property. Viscoelastic fluids which apparently possess this property are, therefore, capable of forming filaments that are appreciably longer than filaments of a Newtonian fluid of the same zero viscosity.

In pursuance of the third objective, the stability of a liquid filament in steady extension (relative to small surface disturbances) has studied. The rheological behavior of the filament fluid has been represented by Noll's Simple Fluid Theory. Constitutive equations which describe the response of a simple fluid to small perturbations superposed on steady extension have been developed. The development rests upon the assumption that the matrix of physical components of the superposed rate of strain consists only of diagonal terms all of which are independent of the radial direction. Physically this assumption corresponds to the case in which the wave length of a typical disturbance-wave is, at all times, much larger than the unperturbed radius of the filament. The resulting constitutive equations are linear in the superposed strain rate and resemble the integral stress-rate of strain relation of linear viscoelasticity. However, in contrast to the latter, the former relations involve several memory functions which depend on the primary steady extension rate "e".

The subsequent stability analysis, subject to several simplifying assumptions, lead to a characteristic equation which determines the stability of the filament. Accordingly, the stability question is primarily dependent on a complex viscosity $\hat{\eta}(\alpha, e)$ which arises from the constitutive equations mentioned above. For a Newtonian fluid, $\hat{\eta}(\alpha, e) = 3\eta_0$ and the results of the analysis indicate that a filament of a Newtonian fluid in steady extension is more stable than the corresponding filament at rest. For a viscoelastic fluid, definite conclusions can not be established since no information is available on the nature of the complex viscosity $\hat{\eta}(\alpha, e)$. Based on some general conjectures which rest, insofar as it is possible, on physical evidence, the present analysis does, however, indicate that a filament of a viscoelastic fluid in steady extension is appreciably more stable than the corresponding filament of a Newtonian fluid and, naturally, more stable than either a viscoelastic or a Newtonian filament at rest. Thus, the theoretical analysis shows that whereas a filament of a viscoelastic fluid at rest is less stable than a Newtonian filament of the same zero viscosity, the opposite effect is more likely to occur when the filament is under tension.

One of the inescapable conclusions of this work is the need for further intensive studies, experimental and theoretical, in the rheology of fluids. Any future progress in this field depends on the existence of an extensive body of experimental data and the availability of constitutive equations which are capable of describing viscoelastic fluids over a wide variety of flow conditions. For many years the bulk of the work in the rheology of fluids has been confined to the study of viscometric flows. Viscometric flows, however, constitute a small class of simple motions and, as such, can not furnish any significant information regarding the behavior of viscoelastic fluids in non-viscometric flows, particularly time-dependent motions. Furthermore, attempts to project the results of viscometric flows upon the behavior of viscoelastic fluids in general have created numerous misconceptions. Thus, for many years it has been a common practice to predict and interpret viscoelastic behavior in processes such as spinning and film extrusion on the basis of viscometric measurements. In view of the vast differences in the behavior of viscoelastic fluids in viscometric flows on the one hand, and in processes such as spinning on the other, the above approach must be considered as totally erroneous. Furthermore, constitutive equations which have been tested in viscometric flows can not be regarded, a priori, as reliable descriptions of viscoelastic fluids in general. This has been clearly demonstrated in the case of the two Oldroyd fluids which have been examined in our study of the spinning process. The present need calls for studies in new flow regimes in which the properties of viscoelastic fluids can be widely manifested and, hence, provide better opportunities to further our understanding of viscoelastic fluids.

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