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THE COLLEGE OF AERONAUTICS CRANFIELD

NON-NEWTONIAN FLOW IN INCOMPRESSIBLE FLUIDS

Part I A general rheological equation of state Part II Some problems in steady flow

by

A. Kaye.



NOTE NO.134 October, 1962

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Non-Newtonian Flow in Incompressible Fluids

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SUMMARY

A rheological equation of state of the form,

 $p_{ij} - p\delta_{ij} = 2 \int_{-\infty}^{t} \left\{ \frac{\partial \Omega}{\partial J_i} S_{ij} - \frac{\partial \Omega}{\partial J_2} S_{ij}^{-1} \right\} dt'$

is proposed for an incompressible material. Ω is a function of J_t and J_2 , the invariants of Cauchy-Green deformation tensor S_{ij} which relates the deformation at the present time t with that at some past time t'. Ω is also a function of t and t'. Some steady state flow problems are solved for a material obeying this equation. It is anticipated that this equation will be of some use in investigating the flow properties of concentrated polymer solutions and polymer melts.

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LIST OF SYMBOLS

p _{ij} , σ _{;j}	stress tensors
p , p'	isotropic pressures
t	current time
t [*]	past time
×i	current position of a general particle in rectangular cartesian co-ordinates
x'i	past position of a general particle in rectangular cartesian co-ordinates
X _i	position of a particle of an elastic body in the unrestrained state
W	stored energy per unit volume of a strained elastic body
C _{ij}	the deformation tensor for an elastic body
I ₁ , I ₂ , I ₃	the invariants of C _{ij}
s _{ij}	the deformation tensor relating the deformation between the past time t^\prime and the current time t of a flowing body
J ₁ , J ₂ , J ₃	the invariants of S_{ij}
Ω	a function of J_1 , J_2 , J_3 and $t - t'$
C'pq	coefficients in the expansion of W in terms of ${\rm I}_1^{}$ and ${\rm I}_2^{}$
S'ij	coefficients in the expansion of $\ \Omega$ in terms of $J_{_1}$ and $J_{_2}$
G	the shear rate
Ni	defined by equation (2.4)
$\mathbf{M}_{\mathbf{i}}$	defined by equation (2.5)
N'i	defined by equation (2.7)
M'i	defined by equation (2.8)
K ₁ , K ₂ , C ₁ , C ₂	constants in the expansion of Ω in the form given by equation (2.20)
nT	the Troughton viscosity

List of Symbols (Continued)

θ _i	convected co-ordinates
ρ	the density of the liquid
z^*	the normal force per unit area
T .	the tangential force per unit area
g	the acceleration due to gravity

Part I. A General Rheological Equation of State

In the phenomenological theory of large elastic deformations in isotropic materials as developed by Rivlin and others the stress-strain relations have the general form,

$$\mathbf{p}_{ij} = \frac{2}{\mathbf{I}_{2}^{\frac{1}{2}}} \left(C_{ij} \left(\frac{\partial W}{\partial \mathbf{I}} + \mathbf{I}_{i} \frac{\partial W}{\partial \mathbf{I}_{2}} \right) - C_{i\alpha} C_{\alpha j} \frac{\partial W}{\partial \mathbf{I}_{2}} + \mathbf{I}_{3} \frac{\partial W}{\partial \mathbf{I}_{3}} \delta_{ij} \right)$$
(1.1)

in which the suffices take the values 1, 2 and 3 with the usual summation convention; p_{1j} are the physical components of the stress tensor and C_{1j} are the components of the Cauchy-Green deformation tensor defined by,

$$C_{ij} = \frac{\partial x_i}{\partial X_{\alpha}} \neq \frac{\partial x_j}{\partial X_{\alpha}}$$

where x_i and X_i are the rectangular Cartesian co-ordinates of a typical particle in the deformed state and the undeformed state respectively. I_1 , I_2 and I_3 are invariants of the tensor C_{ij} defined by the relations

$$I_{1} = C_{\alpha\alpha}$$

$$I_{2} = \frac{1}{2} (C_{\alpha\alpha}^{2} - C_{\alpha\beta} - C_{\beta\alpha})$$

$$I_{3} = \det C_{ij}$$
(1.2)

W is the elastically stored free energy per unit volume expressed as a function of the invariants I_1 , I_2 and I_3 ; δ_{ij} is the unit tensor.

In this paper we shall consider incompressible materials, in which case $I_3 = 1$ for all deformations and W is a function of I_1 and I_2 only. The stress-strain relations may then be written,

$$p_{ij} - p\delta_{ij} = 2 \left(\frac{\partial W}{\partial I_1} C_{ij} + \frac{\partial W}{\partial I_2} (C_{ij} I_1 - C_{ik} C_{kj}) \right)$$
(1.3)

in which p is an arbitrary hydrostatic pressure following from the assumption of incompressibility.

Rivlin (1956) has considered the possible forms of $W(I_1, I_2)$ and shown that, for incompressible isotropic materials, the stored energy function may always be written in the form,

$$W = W(I_1, I_2) = \sum_{p=1}^{\infty} \sum_{q=1}^{\infty} C'_{pq} (I_1 - 3)^p (I_2 - 3)^q$$
(1.4)

with $C'_{00} = 0$. C'_{pq} is a set of constants which defines the elastic properties of the material.

In the kinetic theory of rubber-like elasticity Treloar (1958) derived a specific form for W for an ideal rubber by investigating the properties of a network of idealised molecular chains. A statistical mechanical analysis of this network, involving certain simplifying assumptions, including that of incompressibility, leads to a stored energy function of the form,

$$W = C_{10}'(I - 3)$$

in which $C'_{10} = \frac{1}{2}$ NkT, where N is the number of network chains per unit volume, k is Boltzmann's constant and T is the absolute temperature.

For a material obeying equation (1.5) the stress-strain relations of equation (1.3) become,

$$p_{ij} - p \delta_{ij} = 2 \frac{\partial W}{\partial I} C_{ij}$$

$$= 2 C'_{i0} C_{ij}$$
(1.6)

Lodge (1956) has extended the statistical mechanical model of the ideal rubber in such a way as to produce a model of a liquid which will exhibit visco-elastic effects. This extension is achieved by assuming that the network junction points of the ideal rubber are no longer permanent but have a finite lifetime. The network can then undergo continuous steady deformation and, with various simplifying assumptions, a rheological equation of state for the model is deduced. This has the form,

$$p_{ij} - p\delta_{ij} = \int_{t'=-\infty}^{t} 2S'_{i0}(t-t') \frac{\partial x_i}{\partial x'_{\alpha}} \frac{\partial x_j}{\partial x'_{\alpha}} dt' \qquad (1.7)$$

in which x_i are the rectangular Cartesian co-ordinates of a particle at current time t, and x'_i are the co-ordinates of the particle at some past time t'. $S'_{10}(t - t')$ is a function of the elapsed time (t - t'), which tends to zero as (t - t')tends to infinity. Lodge interprets this function as a lifetime distribution function for the network crosslinks and writes it $\frac{skT}{2} N(t - t')$. The notation $S'_{10}(t - t')$ has been adopted here because equation (1.7) will be regarded as a simple mathematical generalisation of equation (1.6) in which the constant C'_{10} is replaced by the function $S'_{10}(t - t')dt'$ and the co-ordinates X_i by x'_i ; a summation over all time is taken up to the present time t.

It is the purpose of this paper to write down a generalisation of equations (1.3), the stress-strain relations for an isotropic, incompressible elastic solid obtained by a mathematical generalisation formally analogous to that involved in going from equation (1.6) to (1.7). It is expected that this new equation will define the properties of a class of visco-elastic liquids. The properties of such liquids, when subjected to a variety of known flow histories, will be investigated.

The generalisation of equations (1.3) leads to

$$p_{ij} - p\delta_{ij} = 2 \int_{t'=-\infty}^{t} \left(\frac{\partial\Omega}{\partial J_{i}} S_{ij} + \frac{\partial\Omega}{\partial J_{z}} (S_{ij} J_{i} - S_{i\alpha} S_{\alpha j}) \right) dt'$$
(1.8)

where now,

$$S_{ij} = \frac{\partial x_i}{\partial x'_a} \cdot \frac{\partial x_j}{\partial x'_a}$$

(1.9)

(1.5)

and J_1 and J_2 are invariants of the deformation given in terms of S_{ij} by equations similar to equations(1.2).

Also,

Ω

$$= \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} S'_{pq} (J_1 - 3)^p (J_2 - 3)^q \text{ and } S'_{oo} = 0$$
 (1.10)

in which S'_{pq} are functions of (t - t'), which must tend to zero as (t - t') tends to infinity sufficiently quickly to ensure convergence of the integrals in (1.8).

 Ω is written as a function of J_1 and J_2 , the invariants of the deformation tensor S_{ij} . It is the function which characterises the rheological behaviour of the system and it is, in equation (1.10), represented as a summation of a series of functions $S'_{pq}(t - t)$ by analogy with the corresponding expressions in the elasticity theory. The physical significance of Ω is not understood. In the purely elastic case W is known in the sense that it is the phenomenological variable of stored energy per unit volume. In simple cases there is a molecular interpretation of W, for example the kinetic theory of rubber like elasticity predicts W from molecular considerations. However we do not even understand the phenomenological nature of Ω . It is not proposed, therefore, to attempt a discussion of these quantities here; rather will it be assumed that such a general relation as (1.8) is physically permissible and its properties will be investigated.

The rheological equation of state (1.8) can be rewritten in the form,

$$p_{ij} - p'_{\delta_{ij}} = 2 \int_{t'=-\infty}^{t} \left\{ \frac{\partial \Omega}{\partial J_1} S_{ij} - \frac{\partial \Omega}{\partial J_2} S_{ij}^{-1} \right\} dt'$$
(1.11)

where,

 $S_{ij}^{-1} = \frac{\partial x'_{\alpha}}{\partial x_{i}} \qquad \frac{\partial x'_{\alpha}}{\partial x_{j}} \qquad (1.12)$

and,

$$p' = p + 2 \int_{-\infty}^{\infty} J_2 \frac{\partial \Omega}{\partial J_2} dt' \qquad (1.13)$$

The form (1.11) is more convenient for algebraic manipulation than the form (1.8). The proof of this result will be found in Appendix 1. This equation of state will now be investigated for various steady state flow systems.

Part II. Some Problems in Steady Flow

(a) Simple Shearing Flow

Consider the case of steady rectilinear laminar shear flow in which axes are chosen so that the streamlines are parallel to the x, axis and the shearing planes are perpendicular to the x_2 axis. The kinematic equations are then,

$$x_{1} = x_{1}' + G(t - t') x_{2}'$$

$$x_{2} = x_{2}'$$

$$x_{3} = x_{3}'$$
(2.1)

where G is a constant, the shear rate. By evaluating S_{ij} and S_{ij}^{-1} we find,

$$J_{1} = J_{2} = 3 + G^{2}(t - t')^{2}$$

$$J_{3} = 1$$
(2.2.)

and using equation (1.11) we find,

$$p_{11} - p = N_0 + G^2 N_2 - M_0$$

$$p_{22} - p = N_0 - M_0 - G^2 M_2$$

$$p_{33} - p = N_0 - M_0$$

$$p_{21} = (N_1 + M_1) G$$

$$p_{22} = p_2 = 0$$

ŧ

where,

$$N_{i} = 2 \int_{-\infty}^{t} (t - t')^{i} \frac{\partial \Omega}{\partial J_{i}} dt'$$

and,

$$M_{i} = 2 \int_{-\infty} (t - t')^{i} \frac{\partial \Omega}{\partial J_{2}} dt' \qquad (2.5)$$

It is obvious that the equations of motion and continuity are satisfied. It is to be observed that $\Omega(J_1, J_2)$ will be a function of G so that the viscosity, p ai G $= N_1 + M_1$, is not necessarily a constant. However we notice that $p_{22} - p_{33}$, p_{11} , $\frac{p_{21}}{G}$ are all functions of G^2 , in agreement with the general predictions p22 - P11, of Coleman and Noll (1961).

If,

$$\Omega = S'_{10} (J_1 - 3) + S'_{11} (J_2 - 3)$$

(2.6)

(2.3)

(2.4)

then N and M become constants independent of G. These constants will be called N_i' and ${}^{1}\!M_i',$ where,

$$N'_{i} = 2 \int_{-\infty}^{t} S'_{10} (t - t')^{i} dt'$$

$$M'_{i} = 2 \int_{-\infty}^{t} S'_{01} (t - t')^{i} dt'$$
(2.8)

In this case $p_{11} - p_{22}$ and $p_{11} - p_{33}$ become proportional to the square of the shear rate and the viscosity becomes constant, in agreement with results of Markowitz (1962) for low rates of shear.

(b) Flow through straight pipes of arbitrary cross-section

Following the work of Ericksen, (1956), and Oldroyd, (1958), it is of interest to examine the flow of liquid down a pipe of arbitrary cross-section. Consider an infinitely long pipe whose walls are parallel to the x, axis and whose section is given by $F(x_2, x_3) = 0$. Writing the kinematic equations describing the flow as,

$$x_{1} = x_{1}' + (t - t') f (x_{2}', x_{3}')$$

$$x_{2} = x_{2}'$$

$$x_{3} = x_{3}'$$
(2.9)

we shall examine whether a flow of this type, in which the streamlines are parallel to the x_1 direction, is possible for a liquid obeying the rheological equation of state (1.11).

Evaluating
$$S_{ij}$$
 and S_{ij}^{-1} from (1.9) and (1.12) we find,

$$S_{ij} = \begin{pmatrix} 1 + (t - t')^{2} (f_{2}^{2} + f_{3}^{2}) & (t - t')f_{2} & (t - t')f_{3} \\ (t - t')f_{2} & 1 & 0 \\ (t - t')f_{3} & 0 & 1 \end{pmatrix}$$
(2.10)

$$S_{ij}^{-1} = \begin{pmatrix} 1 & -(t - t')f_{2} & -(t - t')f_{3} \\ -(t - t')f_{2} & 1 + f_{2}^{2}(t - t')^{2} & (t - t')^{2}f_{2}f_{3} \\ -(t - t')f_{3} & (t - t')^{2}f_{2}f_{3} & 1 + f_{3}^{2}(t - t')^{2} \end{pmatrix}$$
where,

$$f_{2} = \frac{\partial f(x_{2}, x_{3})}{\partial x_{2}} \qquad f_{3} = \frac{\partial f(x_{2}, x_{3})}{\partial x_{3}}$$
and hence,

W

$$J_{1} = J_{2} = 3 + (t - t')^{2} (f_{2}^{2} + f_{3}^{2})$$
$$J_{3} = 1$$

Substituting
$$S_{ij}$$
 and S_{ij}^{-1} in (1.11) we get,
 $p_{11} - p = N_0 + N_2 (f_2^2 + f_3^2) - M_0$
 $p_{22} - p = N_0 - M_0 - M_2 f_2^2$
 $p_{33} - p = N_0 - M_0 - M_2 f_3^2$
 $p_{21} = (N_1 + M_1)f_2$
 $p_{31} = (N_1 + M_1)f_3$
 $p_{32} = -M_2 f_2 f_3$

The equation of continuity is obviously satisfied. Since, in this case, there are to be no body forces and the flow is steady and rectilinear, the equations of motion which have to be satisfied are,

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$$\frac{\partial p_{ij}}{\partial x_j} = 0$$
 (2.13)

If we first consider the special case of (1.11) in which Ω takes the form (2.6), we find, on inserting (2.12) into (1.11) and noting that N'_i and M'_i are independent of x_i,

$$(N'_{1} + M'_{1}) (f_{22} + f_{33}) = -\frac{\partial p}{\partial x_{1}}$$

$$f_{2}(f_{22} + f_{33}) + f_{2} f_{22} + f_{23} f_{3} = \frac{1}{M'_{2}} \frac{\partial p}{\partial x_{2}}$$

$$f_{3}(f_{22} + f_{33}) + f_{3} f_{33} + f_{23} f_{2} = \frac{1}{M'_{2}} \frac{\partial p}{\partial x_{3}}$$

$$(2.14)$$

These equations are consistent if,

$$f_{22} + f_{33} = \text{constant} = -P \text{(say)}$$

This equation and the boundary condition of no slip, or, mathematically, $f_2 = f_3 = 0$ on $F(x_2, x_3) = 0$, enable $f(x_2, x_3)$ to be determined. $P(N'_1 + M'_1)$ is in fact the pressure gradient down the tube, and $f(x_2, x_3)$ is of course the velocity distribution for a Newtonian liquid of viscosity $(N'_1 + M'_1)$. In other words, we have shown that a liquid for which Ω takes the form given by (2.6) will flow down a tube of arbitrary cross section with the particles of fluid moving in rectilinear paths.

In general, however, on inserting (2.12) into (1.11), and remembering that now $N_{\underline{i}} \text{ and } M_{\underline{i}} \text{ are functions of } x_{\underline{2}} \text{ and } x_{\underline{3}} \text{ , we get,}$

(2.12)

(2.15)

$$-\frac{\partial p'}{\partial x_{1}} = \frac{\partial}{\partial x_{2}} \left[(M_{1} + N_{1})f_{2} \right] + \frac{\partial}{\partial x_{3}} \left[(M_{1} + N_{1})f_{3} \right]$$

$$\frac{\partial p'}{\partial x_{2}} = \frac{\partial}{\partial x_{2}} \left[M_{2}f_{2}^{2} \right] + \frac{\partial}{\partial x_{3}} \left[M_{2}f_{2}f_{3} \right]$$

$$\frac{\partial p'}{\partial x_{3}} = \frac{\partial}{\partial x_{3}} \left[M_{2}f_{3}^{2} \right] + \frac{\partial}{\partial x_{2}} \left[M_{2}f_{2}f_{3} \right] \quad \text{where } p' = p + N_{0} - M_{0}$$

$$(2.16)$$

This set of equations cannot normally be satisfied by some function $f(x_2, x)$. That is, the liquid will not necessarily flow down the tube of arbitrary cross section with rectilinear streamlines. However, for the special case of tubes of circular cross section, it can be shown that the liquid will always flow down the tube with streamlines parallel to the axis of the tube.

(c) Troughton or elongational flow

Consider a state of flow in which a liquid filament is elongated at a constant rate of strain; that is,

$$\frac{dx}{dt} = k_1 x_1, \qquad \frac{dx}{dt} = k_2 x_2, \qquad \frac{dx}{dt} = k_3 x_3 \qquad (2.17)$$

where k_i are constants describing the flow. If now we take $k_1 = a$ and $k_2 = k_3 = -\frac{1}{2}a$ we have simple elongational flow. We have taken $k_1 + k_2 + k_3 = 0$ since this is required by the constant volume condition. The kinematic equations become,

$$x_{1} = x_{1}' e^{a(t-t')} \qquad x_{2} = x_{2}' e^{-\frac{1}{2}a(t-t')} \qquad x_{3} = x_{3}' e^{-\frac{1}{2}a(t-t')}$$
(2.18)
$$J_{1} = e^{2a(t-t')} + 2e^{-a(t-t')} , \qquad J_{2} = e^{-2a(t-t')} + 2e^{a(t-t')}$$

$$J_{3} = 1$$

The equation of state gives,

giving,

$$p_{22} - p_{33} = p_{12} = p_{23} = p_{31} = 0$$

$$p_{11} - p_{22} = \int_{-\infty}^{t} 2 \left(\frac{\partial \Omega}{\partial J_1} \left(e^{2a(t-t')} - e^{-a(t-t')} \right) - \frac{\partial \Omega}{\partial J_2} \left(e^{-2a(t-t')} - e^{a(t-t')} \right) \right) dt'$$
(2.19)

We observe that since Ω is a function of (t - t') the transformation $\tau = t - t'$ proves that $p_1 - p_{22}$ is not a function of t. Let us consider the special case where Ω takes the form,

$$\Omega = C_1 e^{-K_1(t-t')} \left[J_1 - 3 \right] + C_2 e^{-K_2(t-t')} \left[J_2 - 3 \right]$$
(2.20)

where C_1 and C_2 are constants independent of (t - t'). The reason for this is that Lodge (1956) expands S_{10}' in the form,

$$S_{to}' = \sum_{m=1}^{\infty} a_m e^{-\lambda_m (t - t')}$$
 (2.21)

and is able to give a physical meaning to λ_m . A natural extension of (2.21) to the form of Ω given in (1.10) is to take

$$S'_{pq} = \sum_{r=1}^{\infty} b_{pqr} e^{-\lambda_{pqr}(t-t')}$$
 (2.22)

where b and λ_{pqr} are independent of (t - t'). Equation (2.20) represents a form of Ω in which b = C₁, b₀₁₁ = C₂, $\lambda_{101} = K_1$ and $\lambda_{011} = K_2$. All the other b's and λ 's are zero. If $a^{10} < \min(\frac{1}{2}K_1, K_2)$ we find, by evaluating (2.19),

$$p_{11} - p_{22} = \frac{3a. 2 C_1}{(K_1 + a)(K_1 - 2a)} + \frac{3a. 2C_2}{(K_2 + 2a)(K_2 - a)}$$
 (2.23)

If we call $\eta_T = \frac{p_{11} - p_{22}}{a}$, the Troughton viscosity, then

$$\eta_{\rm T} = \frac{3 \frac{2C_1}{K_1}}{\left(1 + \frac{a}{K_1} \right) \left(1 - \frac{2a}{K_1}\right)} + \frac{3 \frac{2C_2}{K_2}}{\left(1 + \frac{2a}{K_2} \right) \left(1 - \frac{a}{K_2}\right)}$$
(2.24)

and we note that, as a tends to zero,

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$$\eta_{\rm T} = 3 \left\{ \frac{2C_1}{K_1^2} + \frac{2C_2}{K_2^2} \right\} = 3.$$
 (shear viscosity)

The form of the two functions involved in η_{T} is shown in Figs. I and II.

We shall now discuss the physical significance of these results. It is well known that elastic liquids readily form filaments of liquid: thus, if a rod is dipped into a polymer solution and then withdrawn, a filament of liquid will be withdrawn with it. Most Newtonian liquids do not have this property, and it seems reasonable therefore that the elastic nature of these liquids may sometimes explain this effect.

Lodge (1960), has investigated the stability of such elongational flow: for a filament which varies in thickness along its length, he has shown that if the elongational viscosity increases sufficiently quickly with rate of strain then a thin section of the filament will decrease in area less rapidly than a thick section. The flow will then obviously be stable.

The above mathematical analysis investigates a simple model for the elongation of a liquid filament: we find that the elongational viscosity rises rapidly with rate of strain, (Figs. 1 and II). It can be shown that certainly somewhere in the range of strain rates $0 < a < \min(\frac{1}{2}K_1, K_2)$ there is a strain rate a_0 above which the elongational flow is stable, in the Lodge sense, and below which it is unstable. Thus the equation of state describes qualitatively the phenomenon of filament formation.

(d) Flow between a rotating cone and a stationary plane

Let the liquid be contained between a horizontal plane and a cone, of semivertical angle $\frac{1}{2}\pi - \alpha$, whose apex touches the plane, whose axis is vertical, and which is rotating with an angular velocity ω . Let the boundary of the liquid be the sphere of radius a, with its centre at the apex of the cone. The shearing laminae are assumed to be cones co-axial with, and with the same vertex as, the rotating cone. The fluid particles are assumed to remain at fixed distances from the apex during the motion. We use the equation of state in the form A9 (see Appendix II).

We take the spherical polar co-ordinates (r, θ, ϕ) as our curvilinear orthogonal set $(\theta_1, \theta_2, \theta_3)$ at time t, and we investigate whether the kinematic equation (2.25) satisfies the equations of motion and continuity.

\mathbf{x}_{1}	=	$r \cos \phi \sin \theta$	$x'_{i} = r \cos(\phi - \tau \theta) \sin \theta$	
х ₂	=	$r \sin \phi \sin \theta$	$x'_{2} = r \sin(\phi - \tau \theta) \sin \theta$	(2.25)
x 3	=	r cos θ	$x'_{3} = r \cos \theta$	

where, $\tau = \frac{\omega}{\alpha} (t - t')$

$$h_{(1)} = 1$$
 $h_{(2)} = r$ $h_{(3)} = r \sin \theta$

where h_(i) is defined by equation A5, Appendix II.

Inserting equations (2.25) into A10 we find,

$$S_{ij} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & r \sin \theta \\ 0 & r \sin \theta & 1 + r^2 \sin^2 \theta \end{pmatrix}$$

$$S_{ij}^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 + r^2 \sin^2 \theta - r \sin \theta \\ 0 & -r \sin \theta & 1 \end{pmatrix}$$
(2.26)
(2.27)

where,

$$J_{1} = J_{2} = 3 + r^{2} \sin^{2} \theta$$

$$J_{3} = 1$$
(2.28)

and we get from A9, (2.26) and (2.27), with the usual notation for the physical components of the stress tensor in polar co-ordinates,

$$(rr) - p = N_0 - M_0$$

$$\begin{pmatrix} \hat{\Theta} \hat{\Theta} \end{pmatrix} - p = N_{O} - M_{O} - \frac{\omega^{2}}{\alpha^{2}} \sin^{2} \hat{\Theta} \cdot M_{2}$$

$$\begin{pmatrix} \hat{\phi} \hat{\phi} \end{pmatrix} - p = N_{O} + \frac{\omega^{2}}{\alpha^{2}} \sin^{2} \hat{\Theta} \cdot N_{2} - M_{O}$$

$$\begin{pmatrix} \hat{\Theta} \hat{\phi} \end{pmatrix} = (N_{1} + M_{1}) \frac{\omega}{\alpha} \sin \theta$$

$$\begin{pmatrix} \hat{\Phi} \hat{\phi} \end{pmatrix} = (\hat{\Phi} \hat{\phi}) = 0$$

$$(2.29)$$

The equation of continuity for incompressible materials is

$$div v = 0$$

where \underline{v} is the velocity of a particle. Using (2.25) we see that this is obviously satisfied.

The equations of motion are,

$$\frac{\partial}{\partial r} (\hat{r}\hat{r}) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{r}\hat{\theta}) + \frac{1}{r\sin\theta} \frac{\partial}{\partial \phi} (\hat{r}\hat{\phi}) + \frac{1}{r} \left[2(\hat{r}\hat{r}) - (\hat{\theta}\hat{\theta}) - (\hat{\phi}\hat{\phi}) + (\hat{r}\hat{\theta} \cot\theta) \right] \\ = -\rho g \cos\theta - \rho r \sin^2\theta \frac{\omega^2}{\alpha^2} \theta^2 \\ \frac{\partial}{\partial r} (\hat{r}\hat{\theta}) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{\theta}\hat{\theta}) + \frac{1}{r\sin\theta} \frac{\partial}{\partial \phi} (\hat{\theta}\phi) + \frac{1}{r} \left[\left((\hat{\theta}\hat{\theta}) - (\hat{\phi}\phi) \right) \cot\theta + 3 (\hat{r}\hat{\theta}) \right] \\ = \rho g \sin\theta - \rho r \sin\theta \cos\theta \frac{\omega^2}{\alpha^2} \theta^2 \\ \frac{\partial}{\partial r} (\hat{r}\phi) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{\theta}\phi) + \frac{1}{r\sin\theta} \frac{\partial}{\partial \phi} (\hat{\phi}\phi) + \frac{1}{r} \left[3 (\hat{r}\phi) + 2 (\hat{\theta}\phi) \cot\theta \right] \\ = 0$$

$$(2.30)$$

where ρ is the density of the liquid.

It can be shown that equations (2.29), when inserted into (2.30), are compatible when $\theta = \frac{1}{2}\pi$, and when inertial forces and body forces are ignored. That is, an approximate solution to equation (2.30), when α is small, may be obtained by putting $\theta = \frac{1}{2}\pi$ and ignoring inertial and body forces. We then find that (2.30) becomes,

$$\frac{\partial p}{\partial r} + \frac{1}{r} \left[M_2 - N_2 \right] \frac{\omega^2}{\alpha^2} = 0$$

$$\frac{1}{r} \frac{\partial p}{\partial \theta} = 0$$

$$\frac{\partial p}{\partial \phi} = 0$$
(2.31)

and hence,

$$p = -\log r (M_2 - N_2) \frac{\omega^2}{\alpha^2} + constant$$
 (2.32)

Using the boundary condition that $\begin{pmatrix} \Lambda \\ rr \end{pmatrix} = 0$ when r = a, we find,

$$\begin{pmatrix} \widehat{\Theta} \Theta \end{pmatrix} = (N_2 - M_2) \frac{\omega^2}{\alpha^2} \log \frac{r}{\alpha} - M_2 \frac{\omega^2}{\alpha^2}$$
 (2.33)

Noting that the normal force Z^* per unit area on the plane is $-(\hat{\theta}\hat{\theta})$ when $\theta = \frac{1}{2}\pi$, we obtain

$$Z^{*} = (N_{2} - M_{2}) \frac{\omega^{2}}{\alpha^{2}} \log \frac{a}{r} + M_{2} \frac{\omega^{2}}{\alpha^{2}}$$

$$(\widehat{\theta}\phi) = (N_{1} + M_{1}) \frac{\omega}{\alpha}$$
(2.34)

also,

If the shear force per unit area on the plane is T, then

$$\Gamma = (\widehat{\theta}\phi)$$
 at $\theta = \pi/2$

and

$$T = (N + M) \frac{\omega}{\alpha} \text{ at } \theta = \pi/2$$
(2.35)

(e) Flow between parallel planes

Let the liquid be sheared between two horizontal parallel planes, with one plane fixed and the other rotating about its normal with a constant angular velocity ω . Let the boundary of the liquid be a cylinder, of radius a, centred on the axis of rotation. If the shearing laminae are assumed to be planes parallel to the fixed plane, and the distance of any particle from the axis of rotation is constant, we may investigate whether the kinematic equations,

$$x_{1} = r \cos \theta \qquad x_{1}' = r \cos (\theta - KZ)$$

$$x_{2} = r \sin \theta \qquad x_{2}' = r \sin (\theta - KZ) \qquad (2.36)$$

$$x_{2} = Z \qquad x_{3}' = Z$$

satisfy the equations of motion and continuity. We use as curvilinear co-ordinates $(\theta_1, \theta_2, \theta_3)$ the cylindrical polar co-ordinates (r, θ, Z) . $K = \frac{\omega}{L}(t - t')$ where L is the distance between the two planes. We note that,

$$h_{(1)} = 1$$
 $h_{(2)} = r$ $h_{(3)} = 1$

where $h_{(i)}$ is defined in equation A5 (Appendix II), and, from equations (2.36) and A10,

$$S_{ij} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 + r^{2}K^{2} & rK \\ 0 & rK & 1 \end{pmatrix}$$

$$S_{ij}^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & -Kr \\ 0 & -rK & 1 + r^{2}K^{2} \end{pmatrix}$$
(2.37)
(2.38)

and,

~

$$J_1 = J_2 = 3 + r^2 K^2, J_3 = 1$$

Equations (2.37), (2.38) and A9 give, using the usual notation for the physical components of the stress tensor in cylindrical polar co-ordinates,

(rr)	-	р	=	N ₀ - M ₀
(źz)	-	р	=	$N_0 - M_0 - \frac{\omega^2 r^2}{L} M_2$
$(\hat{r}\theta)$			æ	$(\mathbf{r}Z) = 0$
($\hat{\theta Z}$)			=	$\frac{r\omega}{L}$ (N ₁ + M ₁)
(00)		р	-	$N_{O} - M_{O} + \frac{\omega^2 r^2}{L^2} N_{I}$

We note that N_i and M_i are functions of r. The equation of continuity is obviously satisfied. The equations of motion are,

$$\frac{\partial}{\partial r} (\hat{r}r) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{r}\theta) + \frac{\partial}{\partial Z} (\hat{r}Z) + \frac{1}{r} \left((\hat{r}r) - (\hat{\theta}\theta) \right) = -\frac{r\omega^2 Z^2}{L} \rho$$

$$\frac{\partial}{\partial r} (\hat{\theta}r) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{\theta}\theta) + \frac{\partial}{\partial Z} (\hat{\theta}Z) + \frac{2(\hat{r}\theta)}{r} = 0 \qquad (2.40)$$

$$\frac{\partial}{\partial r} (\hat{r}Z) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{\theta}Z) + \frac{\partial}{\partial Z} (\hat{Z}Z) + \frac{(\hat{r}Z)}{r} = -g\rho$$

(2.39)

Inserting (2.39) into (2.40) we obtain,

$$\frac{\partial p'}{\partial r} + \frac{1}{r} \left(-\frac{\omega^2}{L^2} r^2 N_2 \right) = -\frac{r \omega^2 Z^2}{L^2} \rho$$

$$\frac{\partial p'}{\partial \theta} = 0$$

$$\frac{\partial p'}{\partial Z} = -\rho g$$
where p' = p + N_0 - M_0
(2.41)

We observe that these equations are inconsistent unless we ignore the inertia term - $\frac{r \omega^2 Z^2}{L^2} \rho$. Ignoring the inertia term and integrating these equations, we get,

$$p' = -\rho_g Z + \frac{\omega^2}{L} \int r N_g dr + constant \qquad (2.42)$$

Using the boundary condition that there is zero surface traction at the surface r = a for all Z, θ , we find that this condition can only be satisfied if we ignore the term $-\rho gZ$ in (2.42). Physically, this is equivalent to considering only very small gaps between the planes. Hence, ignoring the $-\rho gZ$ in (2.42) and using $(\hat{rr}) = 0$ at r = a,

$$p = -N_0 + M_0 + \int_a^r \frac{\omega^2}{L} r N_2 dr$$
 (2.43)

Using (2.43) and (2.39) we find,

$$(ZZ) = \int_{a}^{r} \frac{\omega^2}{L^2} r N_2 dr - \frac{\omega^2 r^2}{L^2} M_2$$
 (2.44)

$$(\widehat{Z}\theta) = \frac{r\omega}{L} (M_1 + N_1)$$
(2.45)

Again the normal force Z^* per unit area on the plane is,

$$Z^{*} = \frac{\omega^{2}r^{2}}{L^{2}} M_{2} + \int_{r}^{\alpha} \frac{\omega^{2}}{L} r N_{2} dr \qquad (2.46)$$

The tangential force T per unit area on the plane is,

$$T = \frac{r\omega}{L} (M_1 + N_2)$$
 (2.47)

(f) Flow between rotating coaxial cylinders

Let the liquid be contained between vertical coaxial cylinders of radii r_1 and r_2 rotating with angular velocities ω_1 and ω_2 . Let us assume that the shearing laminae are cylinders coaxial with the moving cylinders, and that any particle remains at the same height between the cylinders. Taking cylindrical polar co-ordinates (r, θ , Z) for (θ_1 , θ_2 , θ_3), with the Z axis along the axis of the cylinders, we may investigate whether the kinematic equations,

$$x_{1} = r \cos \theta \qquad x_{1}' = r \cos \left[\theta - (t - t') q(r)\right]$$

$$x_{2} = r \sin \theta \qquad x_{2}' = r \sin \left[\theta - (t - t') q(r)\right] \qquad (2.48)$$

$$x_{3} = Z \qquad x_{3}' = Z$$

satisfy the equations of motion and continuity.

From (2.48) and A10 we obtain,

$$S_{ij} = \begin{pmatrix} 1 & r(t - t')q' & 0 \\ r(t - t')q' & 1 + r^{2}(t - t')^{2}q'^{2} & 0 \\ 0 & 0 & 1 \end{pmatrix}$$
(2.49)

$$S_{ij}^{-1} = \begin{pmatrix} 1 + r^{2}(t - t')^{2}q'^{2} & -r(t - t')q' & 0 \\ -r(t - t')q' & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$
(2.50)

where $q' = \frac{dq}{dr}$

also, $J_1 = J_2 = 3 + r^2 (t - t')^2 q'^2 and J_3 = 1.$

Using the equation of state A9 we get, where again the usual notation for the physical components of the stress in cylindrical polar co-ordinates is used,

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$$(\hat{r}\hat{r}) - p = N_0 - M_0 - r^2 q'^2 M_2$$

$$(\hat{\theta}\hat{\theta}) - p = N_0 + r^2 q'^2 N_2 - M_0$$

$$(\hat{Z}\hat{Z}) - p = N_0 - M_0$$

$$(\hat{r}\hat{\theta}) = rq'(N_1 + M_1)$$

$$(\hat{r}\hat{Z}) = (\hat{Z}\hat{\theta}) = 0$$

$$(2.51)$$

where M_{i} and N_{i} are functions of r.

The equations of motion, assuming there are no body forces other than gravity, are,

$$\frac{\partial}{\partial r} (\hat{r}r) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{r}\theta) + \frac{\partial}{\partial Z} (\hat{r}Z) + \frac{1}{r} \left((\hat{r}r) - (\hat{\theta}\theta) \right) = -rq^{2}(r)\rho$$

$$\frac{\partial}{\partial r} (\hat{\theta}r) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{\theta}\theta) + \frac{\partial}{\partial Z} (\hat{\theta}Z) + \frac{2(\hat{r}\theta)}{r} = 0 \qquad (2.52)$$

$$\frac{\partial}{\partial r} (\hat{Z}r) + \frac{1}{r} \frac{\partial}{\partial \theta} (\hat{Z}\theta) + \frac{\partial}{\partial Z} (\hat{Z}Z) + \frac{(\hat{r}Z)}{r} = -g\rho$$

where ρ is the density of the liquid.

Inserting (2.51) into (2.52), we get,

$$\frac{\partial p'}{\partial r} - \frac{\partial}{\partial r} \left[r^2 q'^2 M_2 \right] - \frac{1}{r} \left[r^2 q'^2 \right] \left[N_2 + M_2 \right] = -q^2 \rho r \qquad (2.53)$$

$$\frac{\partial p'}{\partial \theta} + \frac{\partial}{\partial r} \left(r q' (N_1 + M_1) \right) + 2 q' \left(N_1 + M_1 \right) = 0$$
(2.54)

$$\frac{\partial \mathbf{p}'}{\partial Z} = -\rho g \tag{2.55}$$

where $p' = p + N_o - M_o$.

Since $\frac{\partial p'}{\partial \theta} = 0$, equation (2.54) determines q(r) as a function of r when Ω is known. This value of q(r) enables p' to be determined. A knowledge of the boundary conditions will then enable the stress at any point to be determined.

Let us consider the special case when Ω is given by (2.6). Integrating (2.54) and using the boundary conditions that $q(r_1) = \omega_1$ and $q(r_2) = \omega_2$, we find,

$$q(r) = A - \frac{B}{r^2}$$
 hence $q'(r) = \frac{2B}{r^3}$ (2.56)

where

$$A = \frac{r_1^2 \omega_1 - r_2^2 \omega_2}{r_1^2 - r_2^2} \text{ and } B = \frac{r_1^2 r_2^2 (\omega_2 - \omega_1)}{r_2^2 - r_1^2}$$
(2.57)

Using this value of q(r) we can integrate (2.53) to give,

$$p' = \frac{B^2}{r^4} \left(3 M_2' - N_2' \right) - \rho \left(\frac{Ar^2}{2} - 2AB \log r - \frac{B^2}{2r^2} \right) - \rho gZ + \text{constant}$$
(2.58)

where M'_{1} and N'_{1} are given by (2.7) and (2.8).

This gives,

$$(\widehat{ZZ}) = \frac{B^2}{r^4} \left(3M_2' - N_2' \right) + \rho \left(-\frac{Ar^2}{2} + 2AB \log r + \frac{B^2}{2r^2} \right) - \rho gZ + \text{constant}$$
(2.59)

 $Z^* = -(\hat{Z}Z)$ represents the normal forces on a boundary Z = constant which would be required to maintain the assumed state of flow. We see that Z^* is composed of three parts: the first part is due to the non-Newtonian nature of the fluid, the second due to its centripetal acceleration and the third part gives the usual variation of pressure with height in a liquid. For non-Newtonian fluids the first part may be considerably greater than the second part. In this case, if $3M_2 > N_2$ then Z^* is smaller near the centre than it is at the circumference of the gap; hence, if this force were removed, the liquid would tend to fall in the centre and rise at the circumference. However, if $3M_2 < N_2$ the reverse would happen: on removing the force the liquid would tend to rise at the centre and fall at the circumference. The second effect is known as the positive Weissenberg effect and the first as the negative Weissenberg effect.

Discussion

In general, equations of state may be divided into two classes: those which have microrheology as their basis, such as those of Lodge (1956) and Oldroyd (1950), and those which are derived from phenomenological considerations, (Coleman and Noll,1961; Rivlin and Ericson, 1955). The rheological equation of state put forward in this paper belongs mainly to the second class and is an attempt to extend the phenomenological theory of large elastic deformations to a fluid.

The basis of this equation of state is the use of a time dependent function Ω which is a generalisation of W, the stored energy for a purely elastic deformation. The physical significance of Ω is not well understood. Because of the nature of the generalisation process, Ωdt will still have the dimensions of W, energy per unit volume, but must be connected both with the recoverable energy at time t and with the rate of energy dissipation at this time. Thus, even the phenomenological significance of Ω is not clear. The liquid characterised by (1.8) is in a sense a composite relaxing solid composed of many relaxation processes, the elastic modulus for each process being characteristic of a general nonlinear elastic deformation. The energy associated with these processes is gradually dissipated, in some unspecified way, in a fashion determined by the distribution of the relaxation processes and the stress and strain history of the sample. In this way $\Omega~$ is a time dependent distribution of energies among the relaxation processes. Since this equation has been derived from considerations of a thermodynamic stored energy function, it is to be hoped that further investigation will produce a thermodynamic justification for it.

Although the form of Ω is not specified, the advantage of using this generalised form is that it can be altered to fit any experimental results for a large class of liquids. It can be seen, from analysis of simple shear flow, that the normal components of the stress can be varied independently by a suitable choice of Ω , and in this sense all possible experimental results can be analysed in terms of this equation of state. Such a procedure might throw some light on the nature of Ω ; its general form may be specialised to give mathematically simple relations which, if they agree with experiment, may help to place it on a sound physical basis.

Experimental measurements in this field do not, in general, cover a sufficiently wide range of shear rates to enable an unequivocal form of Ω to be chosen. However, an examination of the work of Brodnyan et al (1957) on polyisobutylene solutions shows that a form of Ω which is a function of J, only is sufficient to explain the experimental results. Many more experimental measurements on other liquids over a similar range of shear rates are highly desirable.

A comparison of this equation of state with other equations of state can, at this early stage, only be discussed briefly. Since it includes Lodge's equation of state as a special case, all phenomena which can be explained by his equation can, of course, be equally well explained by this; in addition, this equation will cover the cases in which the normal stresses in simple shear are all different. It would be of interest to enquire whether this equation of state describes a simple fluid in the sense of Coleman and Noll and whether it is included in their general formalism.

Certain types of liquids are obviously not described by this equation of state: liquids which change volume on applying a stress are not included because we have assumed incompressibility; the equation is isotropic and therefore anisotropic liquids are not included; and since the most general form of Ω (2.20) predicts a viscosity which is independent of time, the equation will not describe thixotropic liquids. However, it should be possible, in this last case, to make the S'_{ij} of equation (1.10) functions of t and t' rather than of (t - t'), and

thus to produce a formalism which will describe thixotropy.



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APPENDIX I

Proof that equation (1.8) can be expressed in the form (1.11)

We first note that,

$$S_{ik}^{-1} S_{kj} = \frac{\partial x'_{\alpha}}{\partial x_{i}} \quad \frac{\partial x'_{\alpha}}{\partial x_{k}} \quad \frac{\partial x_{k}}{\partial x'_{\beta}} \quad \frac{\partial x_{i}}{\partial x'_{\beta}} = \delta_{ij}$$
(A.1)

This provides a justification for the use of the notation S_{ij}^{-1} . Let \underline{S} be the matrix whose element belonging to the ith row and jth column is S_{ij} and similarly let the matrix of S_{ij}^{-1} be \underline{S}_{ij}^{-1} . The Cayley-Hamilton theorem states that

$$\underline{\underline{S}}^{3} - J_{1} \underline{\underline{S}}^{2} + J_{2} \underline{\underline{S}} - J_{3} \underline{\underline{I}} = 0$$
 (A.2)

where \underline{I} is the matrix of δ_{ij} .

From A2 we obtain,

$$\underline{S} \quad J_{1} \quad - \quad \underline{S}^{2} = J_{2} \quad \underline{I} \quad - \quad J_{3} \quad \underline{S}^{-1} \tag{A.3}$$

For incompressible materials $J_3 = 1$.

Now by inserting equation A.3 into the equation of state (1.8) we obtain the form (1.11), where p' is given by (1.13).

APPENDIX II

The equation of state in terms of convected co-ordinates

At the current time t the position of the particle can be represented by rectangular Cartesian co-ordinates x_i or by a system of orthogonal curvilinear co-ordinates θ_i where,

$$\mathbf{x}_{i} = \mathbf{x}_{i} \left(\boldsymbol{\theta}_{i} \right) \tag{A.4}$$

and ds is a small element of length given at time t by,

$$ds^{2} = dx_{1}^{2} + dx_{2}^{2} + dx_{3}^{2} = h_{(1)}^{2} d\theta_{1}^{2} + h_{(2)}^{2} d\theta_{2}^{2} + h_{(3)}^{2} d\theta_{3}^{2}$$
(A.5)

We now "name" the particle by the co-ordinates θ_i , and, for all times t', θ_i becomes a convected set of co-ordinates, which are of course only orthogonal at time t. Therefore we must have,

$$\mathbf{x}_{i}^{\prime} = \mathbf{x}_{i}^{\prime} \left(\boldsymbol{\theta}_{i} \right) \tag{A.6}$$

At the time t the physical components of the stress are given by p_{ij} with respect to the rectangular Cartesian axes x_i . The physical components of the stress at the point θ_i , with respect to rectangular Cartesian axes which are locally coincident with the mutually orthogonal lines θ_i = constant, are given by,

$$\sigma_{ij} = \ell_{ip} \ell_{jq} p_{qq}$$
(A.7)

where,

$$\ell_{ip} = h_{(1)} \frac{\partial \theta_{i}}{\partial x_{p}} = \frac{1}{h_{(i)}} \frac{\partial x_{p}}{\partial \theta_{i}}$$
 (A.8)

where i is not summed.

From (A.7), (A.8) and the equations of state (1.11),

$$\sigma_{ij} - \sigma_{ij} = \int_{-\infty}^{t} \left[2 \frac{\partial \Omega}{\partial J_{i}} \left(h_{(i)} h_{(j)} \frac{\partial \theta}{\partial x_{p}} - \frac{\partial \theta}{\partial x_{q}} - \frac{\partial x_{p}}{\partial x_{\alpha}} - \frac{\partial x_{q}}{\partial x_{\alpha}'} \right) - 2 \frac{\partial \Omega}{\partial J_{z}} \left(\frac{1}{h_{(i)}h_{(j)}} - \frac{\partial x_{p}}{\partial \theta_{i}} - \frac{\partial x_{q}}{\partial \theta_{j}} - \frac{\partial x_{\alpha}'}{\partial x_{p}} - \frac{\partial x_{\alpha}'}{\partial x_{q}} \right) dt'$$

hence,

$$\sigma_{ij} - \sigma \delta_{ij} = \int_{-\infty}^{\infty} 2 \left[\frac{\partial \Omega}{\partial J_{i}} \left(h_{(i)} h_{(j)} \frac{\partial \theta_{i}}{\partial x'_{\alpha}} - \frac{\partial \theta_{j}}{\partial x'_{\alpha}} \right) - \frac{\partial \Omega}{\partial J_{z}} \left(\frac{1}{h_{(i)}h_{(j)}} - \frac{\partial x'_{\alpha}}{\partial \theta_{i}} - \frac{\partial x'_{\alpha}}{\partial \theta_{j}} \right) \right] dt'$$

which is the required result.

(A.9)

 \mathbf{S}_{ij} , referred to the orthogonal set of curvilinear co-ordinates, is given by,

$$S_{ij} = h_{(i)} h_{(j)} \frac{\partial \theta_{i}}{\partial x'_{\alpha}} \frac{\partial \theta_{j}}{\partial x'_{\alpha}}$$
$$S_{ij}^{-1} = \frac{1}{h_{(i)} h_{(j)}} \frac{\partial x'_{\alpha}}{\partial \theta_{i}} \frac{\partial x'_{\alpha}}{\partial \theta_{i}}$$

(A.10)



£,