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Orthogonal stagnation flow, a framework for steady extensional flow experiments*)

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With 9 figures

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Nomenclature

a	–	ratio of extension rates $\dot{\varepsilon}_z/\dot{\varepsilon}_y$
A	m^2	cross-sectional area of fluid stream
F	N	force on cross-section of stream
g	m/s^2	gravitational acceleration
h	m	thickness of planar stream, figure 3
l	m	length of planar stream, figure 3
n	–	normal vector to stream surface
p	Pa	pressure, eqs. [19], [20]
r	m	coordinate
R	m	radius of prescribed circular cross-section at position $x = \pm x_0$, eq. [5]
Δt	s	residence time, eq. [10]
t	–	tangential vector on stream surface, eqs. [25], [26]
T	Pa	stress tensor
v	m/s	velocity
$\dot{V}$	m^3/s	volume flow rate
x, y, z	m	principal coordinates
$\dot{\gamma}$	s^{-1}	rate of strain tensor, eq. [3]
$\dot{\varepsilon}_i$	s^{-1}	principal rate of extension
η	Pa s	shear viscosity
τ	Pa	extra stress tensor

Indices

n	normal to stream surface
t	tangential to stream surface
o	reference position in flow field, eq. [4]
x, y, z	direction of coordinates

1. Introduction

Extensional flows are important in many processing operations, for example fiber spinning, foaming, blow molding and flow in die entrances. Extensional flows are also important for fluid characterization. Extensional viscosity should be very sensitive to particle shape according to predictions of suspension theory (1) and to molecular weight according to kinetic theory for polymer solutions (2).

To better understand the response of real fluids in extension it is highly desirable to have available rheometers capable of generating steady extensional flows. Steady extension allows us to measure extensional material functions of a particular fluid which are only a function of the rate of deformation, just as the steady shear viscosity and shear normal stresses are only functions of the shear rate. However, unlike steady simple shear, steady extensional flow is very difficult to generate. The only successful steady extensional flow experiments in the literature to date are for very viscous polymer melts, with $\eta > 10^3$ Pa s. With such high viscosities, solid test methods can be used, usually with the addition of a buoyancy fluid. *Dealy* (3) has recently reviewed these methods. The most successful of these seems to be the vertical tensile system with feedback control developed at BASF by *Münstedt* and *Laun* (4 – 6).

For lower viscosity liquids, a great variety of methods have been tried, for example controlled fiber spinning (7, 8), the ductless siphon or Fano column (9, 10), bubble growth (11, 12), converging flow (13, 14) and the spinning drop (15). In fact, the development of such tests has been one of the most active areas of recent rheology research. For some of these studies, correct measures of extensional viscosity have been obtained for Newtonian fluids (7 – 10, 15). However, in every case it was not possible to achieve steady extensional flow and thus not possible to separate strain rate and time effects for viscoelastic fluids. Some problems in these flows are

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1. initial or entrance conditions dominating the flow,
2. the existence of a free surface which does not generally obey the kinematics of steady extension,
3. changing of the rate of extension along the path lines of fluid elements (inhomogeneous flow),
4. flow instabilities,
5. failure to eliminate shear.

As *Walters* wrote recently: "At the moment there appears to be no practical way of generating a steady extensional flow in the laboratory for mobile liquid systems" (16).

In this paper, we present a new framework for viewing steady extensional flows, that of steady orthogonal stagnation flow. This approach seems to be helpful in dealing with the problems noted above, particularly entrance conditions. First we describe the kinematics of stagnation flow in general, concentrating on two important special cases: axisymmetric and planar. Then we analyse the stress field in these flows, particularly the normal and shear components at stream surfaces. The stagnation flow framework suggests a number of modifications for existing methods and some new extensional tests. These are discussed with particular emphasis on flow through dies with lubricated boundaries. Finally we present some preliminary results for flow of a polyacrylamide solution through a lubricated uniaxial extensional die.

A similar approach as suggested here has been used by *Giesekus* (17, 18) who studied general flows with constant velocity gradients. His studies concentrate on the kinematics of these flows and their verification in a special experiment.

2. Steady orthogonal stagnation flow

All steady extensional flows can be expressed in terms of steady orthogonal stagnation flow, the impingement of two equal coaxial fluid streams, cf. (19, 20). If the two streams are not coaxial and equal, some shear flow will be generated. Figure 1 shows the collision of two coaxial streams of elliptical cross sections.

The interface between the two streams is a plane of symmetry which intersects the axis at the *stagnation point* ($v = 0$). The flow is called orthogonal stagnation flow, since the axis of the streams is orthogonal to the plane of symmetry.

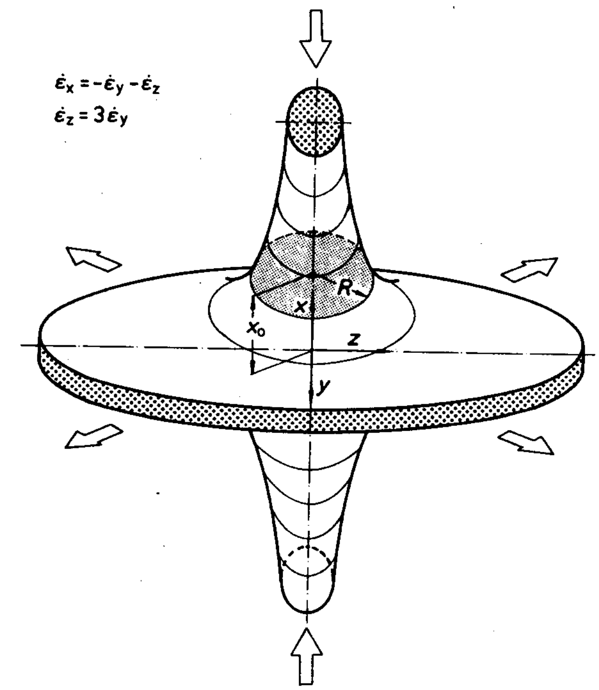

Fig. 1. The collision of two equal, elliptical streams generates general, steady biaxial extension. The large arrows indicate the flow direction. In this case $\dot{V} < 0$. The cross-section at position x_0 is the only circular one. View: tilted around z-axis by 45 degrees

A cartesian coordinate system x, y, z is chosen with the origin at the stagnation point and with its axes in the direction of the principal axes of the rate of strain tensor. As indicated in the figure, the x coordinate lies along the axis of the two streams.

The following analysis is concerned with *steady extensional flow in the entire flow field.* Steady extensional flow of mobile fluids will probably never be achieved completely. However, before trying possible experiments to control the geometry of the extensional region, we first need to know what shape the stream surface should have in an ideal experiment.

2.1. Kinematics

For general steady extensional flow, the velocity field $\boldsymbol{v}$ is given by the Cartesian components

$$(\boldsymbol{v}) = (\dot{\varepsilon}_x x, \dot{\varepsilon}_y y, \dot{\varepsilon}_z z) \qquad [1]$$

where the $\dot{\varepsilon}_i$ are *constant* extension rates. The extension rates may adopt positive or negative values. For an incompressible fluid

$$\dot{\varepsilon}_x = -(\dot{\varepsilon}_y + \dot{\varepsilon}_z)\,. \qquad [2]$$

This is a special case of the flows with constant velocity gradient as investigated by *Giesekus* (17). The vorticity is zero. The components of the rate of strain tensor are

$$(\dot{\gamma}) = \begin{pmatrix} 2\dot{\varepsilon}_x & 0 & 0 \\ 0 & 2\dot{\varepsilon}_y & 0 \\ 0 & 0 & 2\dot{\varepsilon}_z \end{pmatrix}. \qquad [3]$$

The rate of strain is constant in the whole flow field. The fluid elements are elongated at constant rates $\dot{\varepsilon}_x, \dot{\varepsilon}_y, \dot{\varepsilon}_z$ while they move along their path lines, i.e. the flow is steady in a Langrangian sense.

By integrating the components of eq. [1] and by eliminating the time we obtain the path line of a fluid element as the intersect of two cylindrical surfaces

$$(x/x_0)^{1/\dot{\varepsilon}_x} = (y/y_0)^{1/\dot{\varepsilon}_y} = (z/z_0)^{1/\dot{\varepsilon}_z}\,, \qquad [4]$$

where (x_0, y_0, z_0) is a point on the path line.

It is interesting to note that the shape of the entire stream surface is prescribed by prescribing the cross section at *one* position x. For most of the following analysis (an exception is planar extension) it will conveniently be assumed that the fluid streams at positions $x = x_0$ and $x = -x_0$ have a *circular cross-section* of radius

$$R = (y_0^2 + z_0^2)^{1/2}\,. \qquad [5]$$

The shape of the elliptical stream surface then is described by

$$y^2 (x/x_0)^{\frac{2}{1+a}} + z^2 (x/x_0)^{\frac{2a}{1+a}} = R^2\,, \qquad [6]$$

with the ratio of extension rates

$$a = \dot{\varepsilon}_z/\dot{\varepsilon}_y\,. \qquad [7]$$

The fluid streams exhibit an elliptical cross-section at all positions $|x| \neq x_0$. The only exception is axisymmetric extensional flow ($\dot{\varepsilon}_z/\dot{\varepsilon}_y = 1$), where the cross-sections remain circular if a circular cross-section is prescribed at one position x_0. The volume flow rate within the stream surface of eq. [6] is

$$\dot{V} = \pi R^2 v_x(x_0) = \pi R^2 \dot{\varepsilon}_x x_0\,. \qquad [8]$$

Eq. [6] is not valid for the limiting case $x \to 0$, because the stream surface as described in eq. [6] never reaches the plane of symmetry ($x = 0$). A circle on the plane of symmetry with its origin at the stagnation point deforms into an ellipse as the flow continues.

The plane of symmetry is a material plane, i.e. material elements in the plane of symmetry will remain in that plane. Material planes parallel to the plane of symmetry at some distance x remain planar and move with uniform velocity v_x (independently of the position $[y,z]$ of a material element in the plane).

We can also write expressions for the principal strains ε_i which a fluid element experiences passing through a stagnation flow. If an element moves from position x_0 to x then

$$\varepsilon_x = \ln(x/x_0)\,,$$

$$\varepsilon_y = \frac{\dot{\varepsilon}_y}{\dot{\varepsilon}_x}\ln(x/x_0)\,,$$

$$\varepsilon_z = \frac{\dot{\varepsilon}_z}{\dot{\varepsilon}_x}\ln(x/x_0)\,. \qquad [9]$$

The residence time of this material element

$$\Delta t = \frac{\ln(x/x_0)}{\dot{\varepsilon}_x} \qquad [10]$$

is determined by the rate of strain and by the two positions x, x_0.

There are two important special cases of steady stagnation flow: axisymmetric (uniaxial or biaxial) and planar. Each of them is worth being investigated more closely. The governing equations then simplify considerably.

2.1.1. *Kinematics of steady axisymmetric stagnation flow* ($a = 1$)

Consider the collision of two circular streams ($\dot{\varepsilon}_x$ negative, $\dot{\varepsilon}_y = \dot{\varepsilon}_z$). A cylindrical fluid element along the x axis at the inlet plane of the flow will be flattened as it flows toward the stagnation point becoming a large thin disk as $x \to 0$. This is an equal biaxial extension. In this case eq. [6] for the stream surface reduces to

$$(x/x_0)(y^2 + z^2) = R^2\,, \qquad [11]$$

or in cylindrical coordinates (x, r, θ)

$$x r^2 = x_0 R^2\,. \qquad [12]$$

This equation then describes the shape of the two impinging streams. The velocity has the components

$$(v) = (\dot{\varepsilon}_x x, \; -\dot{\varepsilon}_x r/2, 0) \tag{13}$$

and the volumetric flow rate is

$$\dot{V} = \pi R^2 v_x(x_0) = \pi R^2 \dot{\varepsilon}_x x_0 \,. \tag{14}$$

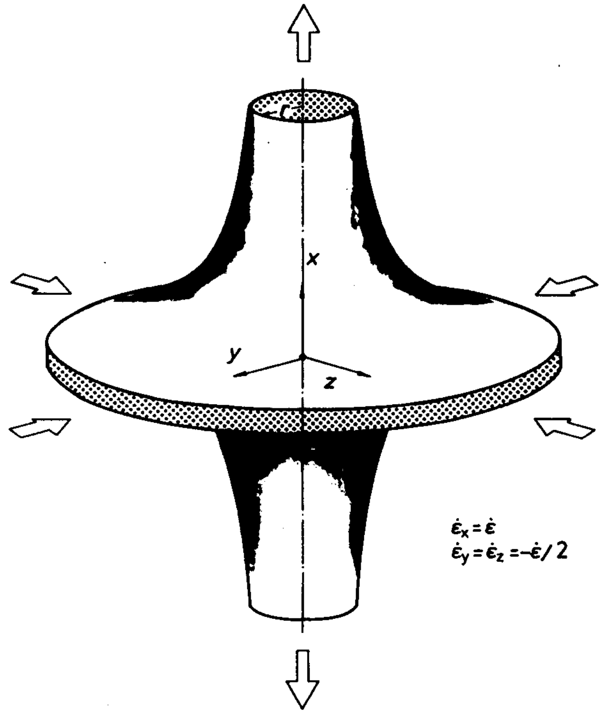

Fig. 2. Steady uniaxial extensional flow generated by flow into a trumpet shaped tube (reverse impingement). The stream surface is axisymmetric. View: tilted forward by 15 degrees.

By reversing the flow rate, the sign of $\dot{\varepsilon}_x$, we obtain uniaxial extension with the same kinematics and flow geometry, see figure 2. However, both cases are interesting rheologically since we expect particles or macromolecules to orient differently in compression and in extension: in uniaxial extension, the macromolecules orient themselves parallel to the *axis* of extension, while in biaxial extension the macromolecules orient themselves parallel to the *plane* of symmetry.

2.1.2. *Kinematics of steady planar extension* ($a = 0$)

Another important special stagnation flow is planar extension (sometimes referred to as "pure shear"). In this case, one of the extension rates is zero, $\dot{\varepsilon}_z = 0$. Planar extension can be achieved by a stagnation flow with an elliptical cross section in which only one axis of the ellipse changes with x. From eq. [6] we obtain the shape of the stream surface

$$(x/x_0)^2 y^2 + z^2 = R^2 \,. \tag{15}$$

Perhaps a more natural way of generating planar extension is the impingement of two rectangular streams as shown in figure 3. The

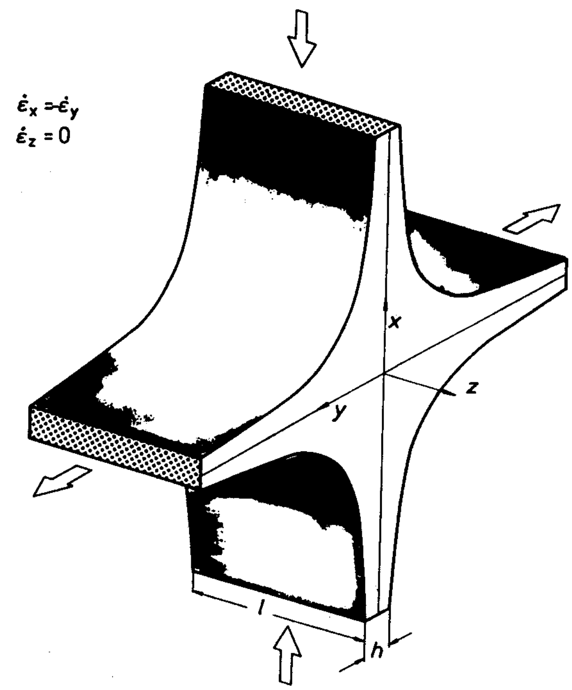

Fig. 3. The impingement of two rectangular streams generates steady planar extensional flow

flow is two-dimensional in a cartesian coordinate system. The shape of the rectangular streams is described by

$$x y = h x_0/2 \,, \tag{16}$$

where h is the thickness of the stream at $|x| = x_0$. The velocity profile is

$$(v) = (\dot{\varepsilon}_x x, \; -\dot{\varepsilon}_x y, 0) \,. \tag{17}$$

The z-axis is a stagnation line ($v = 0$). The volumetric flow rate is

$$\dot{V} = h l \dot{\varepsilon}_x x_0 \,, \tag{18}$$

where l is the length of the stream as indicated in figure 3.

2.2. Stress

In the ideal case of steady extensional flow, the extension rates and the extra stress are constant throughout the fluid. A real experiment, of course, requires an entrance length (like in the measurements of shear rheology with a capillary rheometer). In that entrance length the fluid should be deformed at a constant extension rate until steady extra stress is reached. The magnitude of this entrance region depends on the rheological properties of the fluid. It has to be determined experimentally and shall not be analysed in this study.

When the extra stress is constant throughout the fluid ($\nabla \cdot \tau = \mathbf{0}$), the equations of motion simplify to

$$\rho \dot{\varepsilon}_x^2 x = -\frac{\partial p}{\partial x} + \rho g_x,$$

$$\rho \dot{\varepsilon}_y^2 y = -\frac{\partial p}{\partial y} + \rho g_y,$$

$$\rho \dot{\varepsilon}_z^2 z = -\frac{\partial p}{\partial z} + \rho g_z, \qquad [19]$$

where ρ is the fluid density and the g_i are components of the gravitational acceleration. Solving for the pressure term we obtain

$$p(x,y,z) = p(0,0,0) + \rho(g_x x + g_y y + g_z z) - \frac{\rho}{2}(\dot{\varepsilon}_x^2 x^2 + \dot{\varepsilon}_y^2 y^2 + \dot{\varepsilon}_z^2 z^2). \qquad [20]$$

For fluids of low viscosity the volume forces due to gravity and due to acceleration are important. For molten polymers, however, volume forces are negligible compared to surface forces; the pressure consequently can be assumed to be constant throughout the fluid.

The total force on the cross-section of one of the streams is just the integral of normal stress on the y,z-plane over the cross-sectional area of the stream

$$F_x = \int_A T_{xx} dA. \qquad [21]$$

F_x can readily be evaluated from the normal stress on the particular cross section A.

The boundary conditions are most important when designing an extensional experiment. The normal stress on the stream surface

$$T_{nn} = -p + \tau_{nn} \qquad [22]$$

and the corresponding shear stress T_{nt} depend on the principal stresses T_{xx}, T_{yy}, T_{zz}.

The stress component normal to the curved stream surfaces requires specifying the surface unit normal vector $\boldsymbol{n}$:

$$T_{nn} = \boldsymbol{n} \cdot \boldsymbol{T} \cdot \boldsymbol{n} = n_x^2 T_{xx} + n_y^2 T_{yy} + n_z^2 T_{zz}. \qquad [23]$$

The stress component T_{nn} is the "pressure" that a transducer at position x will read when mounted normal to the stream surface of eq. [6] at the boundary.

In a similar way, we can determine shear stress components at the stream surface

$$T_{nt} = \boldsymbol{n} \cdot \boldsymbol{T} \cdot \boldsymbol{t} \qquad [24]$$

where $\boldsymbol{t}$ stands for any vector in the tangent plane.

Note: The shear stress components T_{nt} are never equal to zero, since the tangent to the stream surface is not a principal plane. Along the curved stream surface, the components T_{nn}, T_{nt} change even if the principal components of the extra stress supposedly stay constant. It will be one of the most difficult tasks of designing steady elongational flow experiments to achieve the stress at the boundary as described in eqs. [23] and [24].

Two unit tangential vectors on the stream surface can be found from eq. [6] by taking the derivative of z with respect to x and y

$$(\boldsymbol{t}_1) = \frac{\left(z, 0, -\frac{a}{1+a}\frac{z^2}{x} - \frac{1}{1+a}\frac{y^2}{x}\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}\right)}{\left[z^2 + \left(\frac{a}{1+a}\frac{z^2}{x} + \frac{1}{1+a}\frac{y^2}{x}\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}\right)^2\right]^{1/2}}, \qquad [25]$$

$$(\boldsymbol{t}_2) = \frac{\left(0, z, -y\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}\right)}{\left[z^2 + y^2\left(\frac{x}{x_0}\right)^{\frac{4(1-a)}{1+a}}\right]^{1/2}}. \qquad [26]$$

These two tangent vectors are in a plane parallel to $y = 0$ and in a plane parallel to $x = 0$, respectively. The two tangent vectors are not perpendicular to each other. It is possible, of course, to choose another pair of tangential vectors for describing the tangent plane at (x, y, z).

The unit normal vector $\boldsymbol{n}$ on the stream surface of eq. [6] is calculated as the cross product of two tangential vectors. The components are

$$(\boldsymbol{n}) = \frac{\left(\frac{a}{1+a}\frac{z^2}{x} + \frac{1}{1+a}\frac{y^2}{x}\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}, y\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}, z\right)}{\left[\left(\frac{a}{1+a}\frac{z^2}{x} + \frac{1}{1+a}\frac{y^2}{x}\left(\frac{x}{x_0}\right)^{\frac{2(1-a)}{1+a}}\right)^2 + y^2\left(\frac{x}{x_0}\right)^{\frac{4(1-a)}{1+a}} + z^2\right]^{1/2}}. \qquad [27]$$

In actual experiments, the pressure transducers will be arranged in the principal planes $y = 0$ and $z = 0$. The components of the normal vector then simplify considerably:

$$(\boldsymbol{n}) = \frac{\left(\frac{a}{1+a}z, 0, x\right)}{\left[\left(\frac{a}{1+a}\right)^2 z^2 + x^2\right]^{1/2}} \quad (\text{for } y = 0), \qquad [28]$$

$$(\boldsymbol{n}) = \frac{\left(\frac{1}{1+a}y, x, 0\right)}{\left[\left(\frac{1}{1+a}\right)^2 y^2 + x^2\right]^{1/2}} \quad (\text{for } z = 0). \qquad [29]$$

The unit tangent vectors in the principal planes are

$$(\boldsymbol{t}) = \frac{\left(x, 0, -\frac{a}{1+a}z\right)}{\left[x^2 + \left(\frac{a}{1+a}\right)^2 z^2\right]^{1/2}} \quad (\text{for } y = 0), \qquad [30]$$

$$(\boldsymbol{t}) = \frac{\left(x, -\frac{1}{1+a}y, 0\right)}{\left[x^2 + \left(\frac{1}{1+a}\right)^2 y^2\right]^{1/2}} \quad (\text{for } z = 0). \qquad [31]$$

These tangent vectors are of special interest for axisymmetric and for planar extension.

2.2.1. *Stress in axisymmetric extension* ($a = 1$)

The above relations reduce considerably for the special case of steady axisymmetrical extensional flow. Letting $\dot{\varepsilon}_y = \dot{\varepsilon}_z$ the cross-sections of the streams are circles of varying radii. The unit normal and the unit tangent on the stream surface in cylindrical coordinates are

$$(\boldsymbol{n}) = \frac{(r, 2x, 0)}{(4x^2 + r^2)^{1/2}}, \quad (\boldsymbol{t}) = \frac{(2x, -r, 0)}{(4x^2 + r^2)^{1/2}} \qquad [32]$$

The normal stress on the stream surface and the tangential stress in flow direction are

$$T_{nn} = \frac{r^2}{4x^2 + r^2}(T_{xx} - T_{rr}) + T_{rr}, \qquad [33]$$

$$T_{nt} = \frac{2rx}{4x^2 + r^2}(T_{xx} - T_{rr}). \qquad [34]$$

At large x, the normal stress T_{nn} approaches T_{rr} while at small x it goes to T_{xx}. The shear stress in the tangent plane vanishes at large x or at large r; it is most pronounced, when r and x are of the same order of magnitude.

In the region of fully developed flow, the principal normal stress differences will be constant and a differential wall pressure measurement at two positions, x_1 and x_2, will give

$$\begin{aligned} T_{nn_1} - T_{nn_2} &= (T_{xx} - T_{rr}) \\ &\cdot \left(\frac{1}{1 + 4x_1^3/C} - \frac{1}{1 + 4x_2^3/C}\right) \\ &- \rho g_x (x_1 - x_2) \\ &+ \rho \dot{\varepsilon}_x^2 \left[\frac{x_1^2 - x_2^2}{2} + \frac{C}{8}\left(\frac{1}{x_1} - \frac{1}{x_2}\right)\right] \end{aligned} \qquad [35]$$

with

$C = x_0 R^2$.

For uniaxial extension (or equal biaxial extension), the extensional viscosity is defined as

$$\eta_E = \frac{T_{xx} - T_{rr}}{\dot{\varepsilon}_x}. \quad [36]$$

Thus with the differential pressure at known positions the extensional viscosity can be evaluated.

2.2.2. Stress in planar extension ($a = 0$)

The unit normal and the unit tangential vector on the stream surface simplify to

$$(\boldsymbol{n}) = \frac{(y,x,0)}{(x^2 + y^2)^{1/2}}; \quad (\boldsymbol{t}) = \frac{(x,-y,0)}{(x^2 + y^2)^{1/2}}. \quad [37]$$

The normal stress T_{nn} on the stream surface and the tangential stress T_{nt} can be evaluated from eq. [37] together with eqs. [23] and [24]:

$$T_{nn} = \frac{y^2}{x^2 + y^2}(T_{xx} - T_{yy}) + T_{yy}, \quad [38]$$

$$T_{nt} = \frac{xy}{x^2 + y^2}(T_{xx} - T_{yy}). \quad [39]$$

The normal stress on the flat surface on the sides of the rectangular streams is the principal stress T_{zz}. Thus, if we succeed in measuring two pressures, one on the curved surface of the stream and one on the flat side, both at the same distance x above the plane of symmetry, we will obtain a combination of the two normal stress differences for planar extension:

$$T_{nn} - T_{zz} = \frac{y^2}{x^2 + y^2}(T_{xx} - T_{yy}) + (T_{yy} - T_{zz}). \quad [40]$$

These divided by the extension rate can be used to calculate the two "planar extensional viscosities". The geometry term in eq. [40] indicates how much of the first normal stress will be measured by the pressure transducers. At large x, near the inlet in figure 3, nearly all of $T_{yy} - T_{zz}$ will be recorded, near the outlet very little.

Note that an additional pressure term from eq. [20] enters eq. [40], if the normal stress T_{zz} is not measured next to the stream surface (xy = const.). This would be the case for measurements in the plane of symmetry ($y = 0$), for instance.

3. Extensional geometries

The framework of stagnation flow given above can help us to modify some current experiments or devise new ones to achieve steady extension. The principle advantage of impinging two streams together (at positive or negative flow rate) rather than stretching a single stream seems to be in the entrance and exit conditions of the flow. Stagnation provides a smoother transition from a necessary delivery system to the desired pure extension. Thus for a given size apparatus more of the flow will be at steady extension and larger extensional strains can be developed, eq. [9]. This is, of course, still contingent on solving the other problems with extensional experiments described in the introduction. Below we suggest in a very qualitative way some possible designs motivated by the stagnation flow framework.

3.1. Free surface

In the current fiber spinning experiments the entrance condition is usually that of shear flow from the delivery tube followed by extrudate swell. It seems that this entrance condition could be improved by the design sketched in figure 4. Here test fluid is extruded inward from a ring

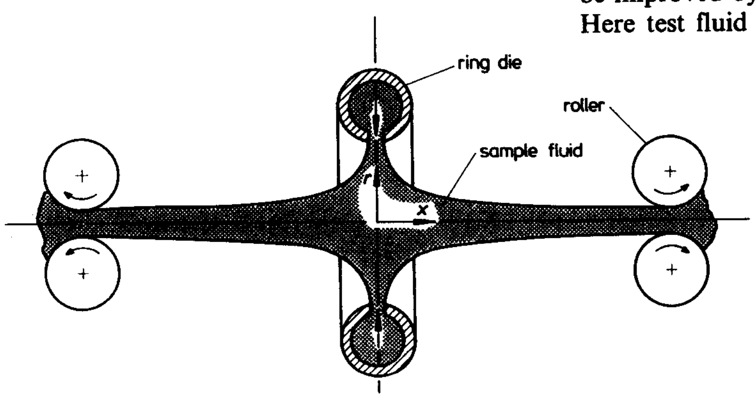

Fig. 4. Spinning in two directions from a ring die to provide large extensional strain

die and drawn off perpendicularly in opposite directions as two cylindrical streams. In this case, one can expect the velocity profile to be closer to uniaxial extension than in the common fiber spinning design. The shear flow from the delivery channel should by comparison be less disruptive. It may be possible to balance gravity with a buoyancy fluid.

A second extensional flow experiment with free surface is uniaxial extension in a ductless syphon [9, 10]. Comparison with the velocity field of stagnation flow shows that the entrance and the exit conditions are not satisfying: At the entrance of the extensional flow region, there is no stagnation point generated, and at the exit, the velocity approaches that of pipe flow.

The stress at the free surface is different from what is prescribed by eqs. [33] and [34]. At a free surface, the shear stress must be zero and the normal stress T_{nn} must be balanced by surface tension. In order to meet these conditions the extra stress might not be homogeneous throughout the fluid.

Another significant problem with these free surface experiments is that, due to surface tension and gravity, the velocity field and the surface shape do not follow eqs. [1] and [6]. In some cases the extension rate increases and in others it decreases along the filament (7–10), i.e. the flow then is not steady in a Langrangian sense. Thus it seems desirable to look at stagnation flow experiments that somehow can be confined between liquid or solid surfaces.

3.2. Stream in a sea of fluid

Frank, Keller, and *Mackley* (21, 22) achieve large extensions of macromolecules in solution by letting two opposed coaxial fluid jets meet in a sea of fluid. The jets come from two coaxial pipes (or slits, respectively). The parabolic velocity field of pipe flow has to rearrange into the velocity field of stagnation flow. Most of the fluid is subjected to shear *and* extensional deformations. Exceptions are the fluid elements moving along the axis of symmetry and moving in the plane of symmetry: they are subjected to pure extensional flow, but at changing rates of extension along their paths. Only in the immediate neighbourhood of the stagnation point, the flow becomes steady extension.

The velocity field of eq. [1] as prescribed by steady extension requires velocities which increase with the distance from the stagnation point. This requirement is in contradiction of flow in a sea of fluid, where the velocity reduces to zero in a distance.

3.3. Moving surfaces

Taylor (23), *Giesekus* (18), and recently *Mackley* (24) used the arrangement of four cylindrical rollers shown in figure 5 to produce planar stagnation flow in a Newtonian fluid. *Parlato,* as communicated in (13), reported that for some

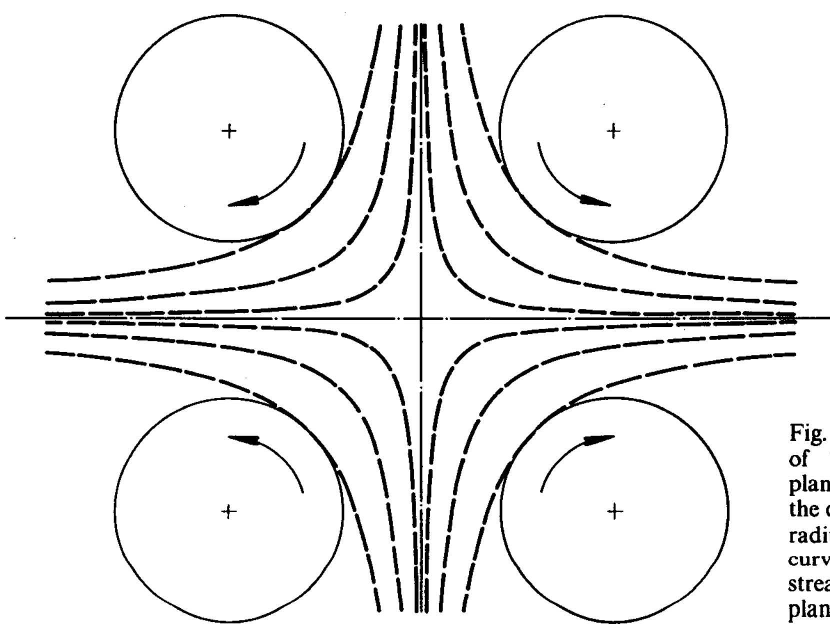

Fig. 5. The four roller experiment of *Taylor* (23). This produces planar stagnation flow only near the center. In the picture the roller radius is chosen to be a radius of curvature of one of the hyperbolic stream lines (dashed) of steady planar extension

viscoelastic fluids the rollers could not pump the fluid. We see that a roller of radius r only locally approximates the desired hyperbolic stream surface $xy = \text{const}$. With flexible belts it would be possible to better approximate the shape. However a major problem is that a continuous belt must move at constant velocity, whereas eq. [1] requires a changing velocity along the surface. Measurements of pressure through such a belt would also be difficult.

Another approach might be to construct a surface from a row of many small rollers, each of which can assume the velocity necessary for planar extensional flow. The experimental problems of roller friction, sealing the spaces between the rollers, and measuring pressures in such an apparatus seem to be formidable.

3.4. Porous walls

It is possible, in principle, to achieve a steady stagnation flow by controlled removal (or injection) of fluid through porous walls. Such an idea is schematically shown in figure 6. From kinematics we know that the removal rate per unit area has to be constant along the entire inner wall of the pipe. Here we suggest control of fluid removal by varying the pressure drop through the holes, lower drop near the center. The design is relatively simple, however in this case relative flow rates would also be controlled by the viscosity shear rate relation for the test fluid. Flow rate could also be controlled by varying diameter or concentration of pores. In either case the sections of solid wall will contribute shear to the flow.

Note that for flow with negligible inertia and gravity the pressure is constant throughout the fluid. In this case, the distribution of pores has to be uniform along the boundary.

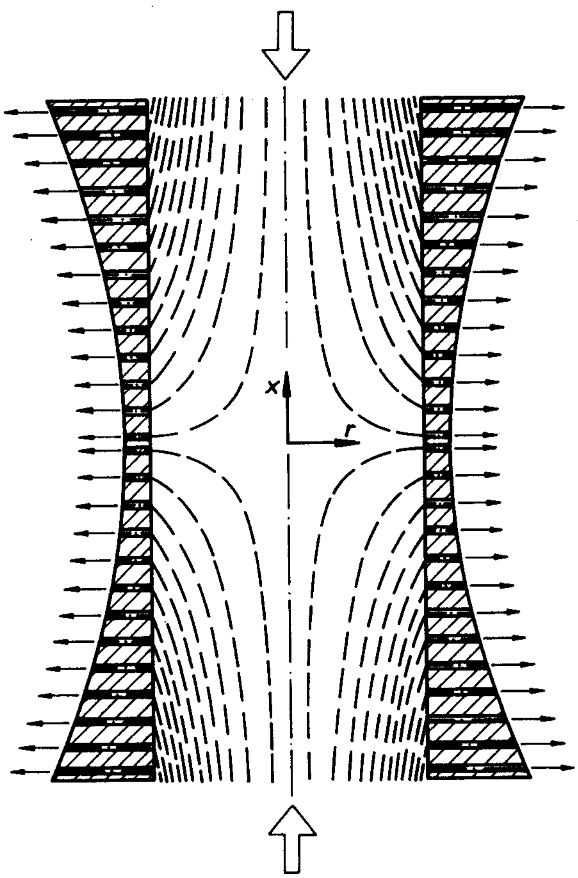

Fig. 6. Approximation of axisymmetric stagnation flow by flow into a porous tube with a gradient of pores length

3.5. Lubricated surfaces

An approach which looks more promising the above concepts is to provide some means of promoting slip at solid boundaries. Some polymer melts such as polyvinylchloride (25) and polytetrafluorethylene (26) are known to slip at die surfaces. In many cases, a low molecular weight insoluble species is added as a "lubricant" or processing aid to promote slip. *Shaw* (27) reports that a coating of silicone grease on the wall of a conical die reduced pressure drop for polyethylene melts. In these later cases we clearly have two-phase flow with a relatively low viscosity phase in a thin layer at the wall carrying most of the shear stress. Very recently *Everage* and *Ballman* (28) have reported using a continuously lubricated die with a converging circular cross-section for uniaxial extensional flow measurements.

This concept seems to have potential for a wide variety of experiments. If suitable lubricants and injection means can be found then solid dies with the shapes shown in figures 1 to 3 can be made to produce any type of steady extension flow. Another possible design is to impact a stream onto a lubricated plate as indicated in figure 7. The shape of the surface probably has to be prescribed by a lubricated die.

If the lubricant layer is relatively thin and of much lower viscosity than the test fluid, then the flow in one of these lubricated die geometries will indeed be a stagnation flow and described by the equations developed above. The exten-

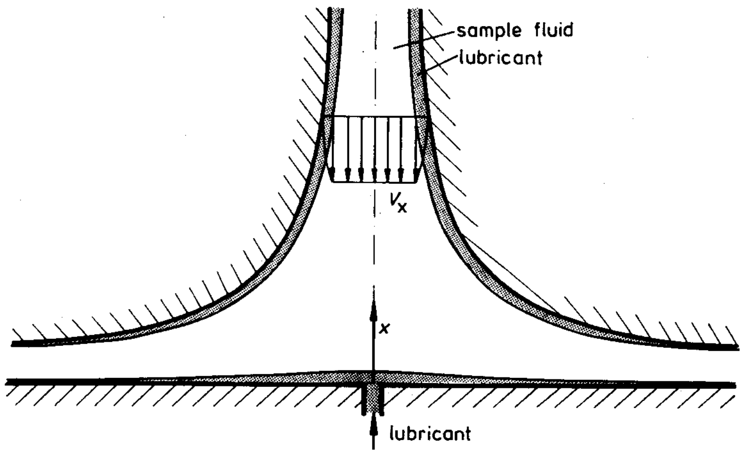

Fig. 7. Collision of an axisymmetric stream with a lubricated plate. The stream is contained in a lubricated die

sional stresses can be evaluated from pressure taps in the solid surfaces reading through the lubricant layer or the total thrust on a plate such as in figure 7. The pressure taps will measure the component of the stress field inside the flow acting normal to the solid surface, T_{nn}.

4. Conclusions and preliminary results with a lubricated die

The description of the ideal extensional experiment for measuring viscosities of steady extension demonstrates the opportunities and the disadvantages of known experiments. The most pronounced deviations arise from insuf-

Fig. 8. Material surfaces deforming in converging die flow with no -slip wall condition. Fluid: 3.5% polyacrylamide solution in H_2O. Flow rate 6.85 cm^3/s. Die contour: $x r^2 = 8.7$ cm^3

ficient initial and boundary conditions for the velocity field and for the stress.

Of all the concepts suggested above we find the lubricated dies most attractive. However there are considerable constraints on the lubricant. The boundary conditions on the lubricant layer are:

1. constant lubricant flow rate,
2. lubricant velocity zero at the die surface,
3. lubricant velocity equal to the test fluid velocity at the interface,
4. shear stress balance across the interface,
5. normal stress balance across the interface.

The problem, of course, is to satisfy all these boundary conditions and still achieve the desired steady extensional flow. The choice of lubricant viscosity and flow rate and even possibly the die shape will be an optimization problem complicated by potential interface instability. It may be best to remove surface tension effects at the interface by using a lubricant soluble in the test fluid. Residence time can be made short enough so that the test fluid will be essentially unchanged during the extension.

We are presently carrying out such lubricated die experiments (29). We have constructed an axisymmetric die from plexiglass which follows the xr^2 = const. shape. We used a 3.5% polyacrylamide (Cynamer P-250) solution in water, zero viscosity $\eta_0 = 100$ Pa s. Velocities were measured by hydrogen bubbles as described by *Schraub* et al. (30) using a 25 μm diameter tungsten wire with 100 V DC.

Figure 8 shows velocity profiles for a 6.85 cm^3/s flow rate of polymer solution without lubricant. Note the very severe velocity gradient and the apparently zero velocity at the wall. In figure 9 the same polymer flow rate was used but with approximately 3.42 cm^3/s water flow rate introduced at the wall. We found that with an insoluble lubricant such as silicone oil the flow was unstable; the oil would not uniformly wet the walls. In figure 9 the water layer is the black layer particularely apparent on the left side of the flow. The velocity profiles are nearly flat.

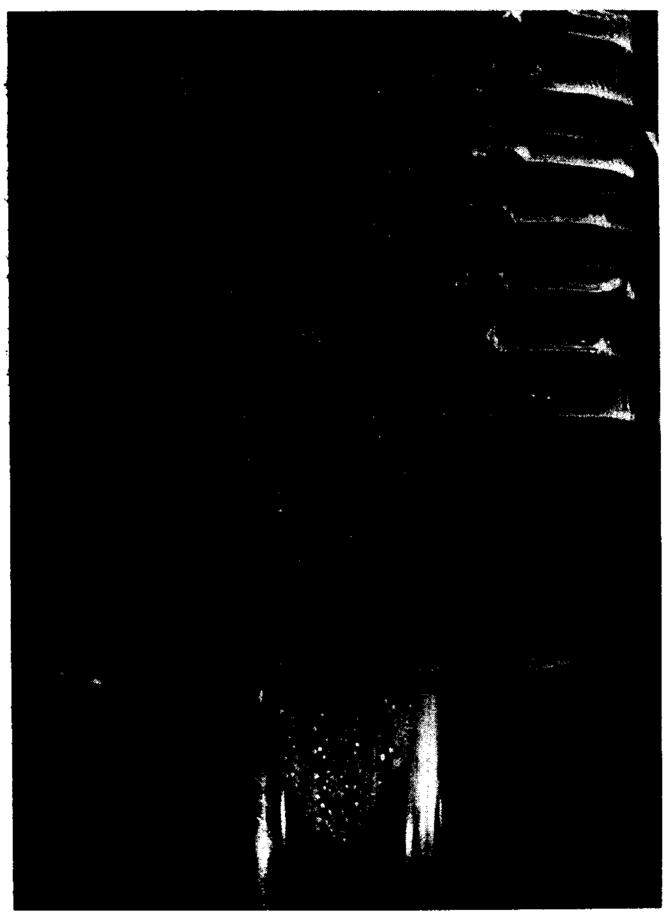

Fig. 9. Material surfaces deforming in converging die flow with lubricated walls. Fluids: polyacrylamide solution with H_2O as lubricant. Flow rates: 6.85 cm^3/s and 3.42 cm^3/s

Distortion of the profiles near the exit may be due to a valve at the exit. Wall pressure drop in the lubricated case was much less than for the non-slip flow. A more complete account of these experimental results along with an analysis of flow in the lubricant layer has been presented elsewhere (29).

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Summary

A new framework for viewing steady extensional flow is presented: Steady orthogonal stagnation flow is an ideal which one should strive to reach in an actual experiment. The kinematics and the stress boundary conditions are developed for stagnation flow in general and in two special cases: the impingement of two circular streams and of two planar sheets. Contemplating this ideal clarifies the advantages and disadvantages of current experiments, thereby pointing the way towards new experiments; a number are suggested. Axisymmetric (with $\dot{\varepsilon}_x > 0$) and planar stagnation flow within a lubricated die look particularly promising. Some preliminary experimental results are given for uniaxial extension of a polyacrylamide solution in a water lubricated die.

Zusammenfassung

Es werden neue Leitlinien zur Beurteilung von stationären Dehnströmungs-Experimenten vorgeschlagen: Die stationäre orthogonale Staupunktströmung wird als ein Ideal herausgestellt, das man in Experimenten zu erreichen versuchen sollte. Die Kinematik und die Randbedingung für das Spannungsfeld werden für allgemeine Staupunktströmungen hergeleitet und auf zwei Sonderfälle angewendet: die axialsymmetrische und die ebene Staupunktströmung. Der Vergleich dieser idealen Strömung mit wirklichen Experimenten verdeutlicht deren jeweilige Vor- und Nachteile und weist so auf neue Experimente hin. Mehrere neue Experimente werden vorgeschlagen. Die axialsymmetrisch nach außen gerichtete und die ebene Staupunktströmung in Düsen mit geschmierten Wandungen erscheinen besonders erfolgversprechend. Einige vorläufige experimentelle Ergebnisse für die einachsige Dehnströmung einer Polyacrylamidlösung in einer mit Wasser geschmierten Düse werden vorgestellt.

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