

Review of converging flow methods for determining the extensional flow behaviour of polymer melts

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ABSTRACT

In many forming processes the polymer melt undergoes predominantly extensional deformation, for example in blow moulding and film blowing. Furthermore, the extensional flow behaviour of melts can differ significantly from their shear flow behaviour. Thus extensional flow measurement methods are necessary to characterise most appropriately the melt behaviour. Such data can be used for the development and selection of materials and for improving processing.

Converging or contraction flow methods can be used to assess the extensional flow behaviour of polymer melts. However, there are several methods available that can be used to interpret the raw entrance pressure drop data. A critical review of converging flow methods for determining the extensional viscoelastic behaviour of polymer melts is presented. The converging flow models, their use and comparison with other methods are reviewed.

The Cogswell model is the most favoured, as indicated by usage in the literature. Good agreement with tensile test methods has been reported, although improved agreement was obtained by modifying the model to incorporate a rate dependent extensional viscosity term and by applying a correction for non-Newtonian shear flow effects (Rabinowitsch correction).

This information will help to establish good practice in use of such models so that the technology can be most appropriately, easily and effectively used by industry.

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FIGURE

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1. INTRODUCTION

In many plastics forming processes the melt undergoes predominantly extensional deformation, for example in blow moulding, fibre spinning, film blowing and vacuum forming (1-3). Furthermore, the extensional flow behaviour of polymer melts can differ significantly from their shear flow behaviour. For example, extensional viscosity values may be an order of magnitude or more higher than shear viscosity values and may also exhibit different trends, for example strain hardening as opposed to shear thinning. Thus extensional flow measurement methods are necessary to characterise most appropriately the melt behaviour. Such data can be used for the development and selection of materials and for improving processing.

Extensional flow also occurs in injection moulding, for example in the nozzle of the injection moulding machine and in passing from the runner to the gate of the moulding. To predict the flow of material in such regions, an ability to characterise the behaviour of the material in extensional flow is required.

One approach for determining the extensional flow behaviour of polymer melts is by the measurement of the resistance to flow in a contraction¹, Figure 1. A contraction or converging flow is, by definition, a type of extensional flow. It is expected therefore that extensional viscosity might be obtained from the measurement of pressure drop and flow rate in converging flows. The approach of using converging flow methods is appealing as experimental data are easily obtained from capillary extrusion rheometry testing - see Figure 1 and Section 3.1 for details. Thus both shear and extensional viscosity values can be obtained from the same tests.

However, there are various contraction or converging flow models that are used to analyse the raw experimental data to determine the extensional flow behaviour of polymers. The questions: "Which converging flow model is best suited for the application?", and "How valid are the results obtained from the various models?" remain largely unanswered. A critical review of converging or contraction flow methods for determining the extensional viscoelastic behaviour of polymer melts is therefore presented to provide guidance to answering these questions. This will help establish the best practice in use of such models for determining extensional flow behaviour so that the technology can most appropriately, easily and effectively be used by industry.

This review was carried out as part of the project on measurement of the extensional viscoelastic properties of polymers (project reference MMP11). This project is one of a number of projects funded by the Engineering, Automotive and Metals Division (EAM) of the Department of Trade and Industry. The project addresses the development of methods for characterising the extensional viscoelastic behaviour of polymer melts. The specific objective of the project that this report addresses is:

- To review existing methods for measuring the parameters that describe the viscoelastic behaviour of polymeric materials under stress fields that are predominantly extensional.

This report will be followed by reports on an assessment of the sensitivity of the predictions of selected converging flow models to the input data and on the findings of an intercomparison of extensional viscoelastic measurement methods, including converging flow methods.

¹ A review of stretching methods for the measurement of extensional viscoelastic properties is presented elsewhere (4).

This review commences with comments on extensional viscoelasticity measurement methods indicating the principle difficulties of such measurements and the intercomparison of results from different techniques [Section 2]. This is followed by a description of: the basic principles of converging flow measurements [Section 3.1], the various converging flow models presented in the literature [Section 3.2] and the assumptions typically made in interpreting converging flow measurements as extensional viscoelastic properties [Sections 3.3 and 3.4]. The more popular converging flow models are described in greater detail [Section 3.5 and Appendices] and comparisons with other techniques are reviewed [Sections 3.6].

2. EXTENSIONAL VISCOELASTICITY MEASUREMENTS

This review is of converging or contraction flow methods for extensional viscoelastic measurements of polymer melts. Alternative methods in which a specimen is clamped at each end and then stretched were reviewed in an earlier report as part of the project MMP11 (4).

A significant contribution to the understanding of extensional viscoelasticity measurements, albeit specifically addressing the measurement of polymer solutions, was made through recent international collaborations. Results of measurements of the shear and extensional viscoelastic properties of three polymeric solutions M1, A1 and S1 were reported by various researchers:

- M1 - 0.244% polyisobutylene in a mixed solvent of 7% kerosene in polybutene, exhibiting constant shear viscosity with a dominant relaxation time of 0.3 s (references 5 (26 papers) and 6);
- A1 - 2% polyisobutylene in a mixture of cis- and trans-decalin, exhibiting shear thinning with a dominant relaxation time of 5 s (references 6 and 7); and
- S1 - 2.5% polyisobutylene in a mixture of decalin and polybutene, exhibiting shear thinning with a dominant relaxation time similar to M1 (reference 8 (7 papers)).

The experimental methods that were used are described in the various papers and also by James et al (9) and Gupta et al (10). Comparison of results for the polymeric solutions A1 (7) and M1 (9) clearly indicated substantial differences in extensional properties obtained using the different methods: the extensional viscosity varied by three decades over the two decades of strain rate with results presented over virtually the entire region. Petrie (11) commented that the discrepancy was because "the experimental methods were not reproducing the conditions equivalent to the formal definition of extensional viscosity" - i.e. the tests generated flow fields that were not well approximated by the mathematical descriptions used to interpret the experimental data. James et al (9) commented that the rheometers used in the study of the fluid M1 should be viewed as yielding "*an* extensional viscosity rather than *the* extensional viscosity". Recently, Ferguson et al (12, 13) argued that transient extensional viscosity values (λ_E^+) should be presented as a function of both strain (ϵ) and time (t). They demonstrated that a surface of the 3-D plot $\lambda_E^+ = \lambda_E^+(\epsilon, t)$ for polyisobutylene could be determined on which lay the results of individual tests of different strain rate - time and thus strain - time profiles. However, such a plot does not take into account the strain history of a test, for example at time $t_1 > 0$ two tests may have the same strain but different strain rates as may be the case where one method has a constant strain rate and the other a decreasing strain rate. The uniqueness of the surface is therefore questioned. Although this approach is perhaps a step in the right direction the effect of strain and strain rate history on results needs to be investigated further.

This volume of work on polymeric solutions has clearly highlighted some of the difficulties facing the measurement of extensional flow properties. The method of converging flow analysis is now considered in more detail.

3. CONVERGING FLOW METHODS FOR EXTENSIONAL VISCOELASTICITY MEASUREMENTS

3.1. THE CONVERGING FLOW MEASUREMENT METHOD

The basic principle behind converging flow measurements is that the pressure drop in a contraction flow is due to both the shear and extensional flow properties of the material. Thus given the pressure drop - flow rate relationship for a specified geometry and the shear flow behaviour of the material then the extensional flow behaviour can be deduced, assuming a suitable relationship relating these three components. It is this last stage of what constitutes a "suitable relationship" that is the main topic of this report.

Data for the determination of extensional viscosity values can be obtained from, for example, capillary extrusion rheometry, Figure 1, following ISO 11443: Plastics - Determination of the fluidity of plastics using capillary and slit die rheometers (14). In using a capillary rheometer the pressure upstream of the die entrance is measured. By using two dies of different length but of the same diameter the pressure drops in (i) the converging region upstream of the die and (ii) along the die length (shear flow only) can be separately determined following the approach initially proposed by Bagley (15). Alternatively, a "zero" length die can be used to determine the entrance pressure drop. However, shear viscosity data for the material are also required in applying most of the converging flow models thus requiring measurements using a die of at least one other length.

The data typically required for converging flow analysis are entrance pressure drop versus flow rate for a given geometry and shear viscosity versus shear rate, all at the same test temperature. The geometry required depends on the model that is to be used: it is typically either a right-angled entry geometry or a conical or profiled entry geometry that can be lubricated, if desired, to eliminate shearing forces. Additional information, for example the streamlines obtained through flow visualisation, may also be used to validate assumptions or compare with predictions.

3.2. CONVERGING FLOW MODELS

The subject of using converging flow analysis to determine extensional viscosity data was reviewed by Cogswell (16) in 1978 in the infancy of interest in the use of such an approach. More recently, Binding (17) and Gibson (18) presented their respective views of the use of converging flow analysis, each presenting their own models in detail.

Numerous models for determining extensional viscosity from entrance pressure drop measurements of polymer melts and solutions have been presented in the literature. The salient features of these models are summarised in Table 1. Further application of these models is presented in Table 2. The models can be sub-divided into the categories of constrained flow (19-31) or free convergence flow (22, 32-38, 83). Constrained flow is defined as where the entry to the die is profiled thereby preventing the polymer from forming its own "free convergence" flow pattern into the die (Figure 1). Free convergence models are indicated

in Table 1 as having a 90° entry geometry². However, an entry flow may be of free convergence type when a conical or profiled entry geometry is used if the material's converging flow boundary is contained within that of the profiled boundary. This may occur when using large entry angle geometries. Free convergence models may be used to predict the position of the converging flow boundary for comparison with experimental data or numerical modelling. Constrained flows may also be lubricated using, for example, a lower viscosity polymer or oil (24, 26-28, 30, 39). Even when the flow is not lubricated the assumption that shear forces can be ignored is made in several of the models (26, 29, 32, 33, 35, 83). The use of a lubricated hyperbolic profile in the converging region has the benefit that it results in a flow field of uniform strain rate over the profiled region (24-26, 40-42). However, this approach is not without its problems, in particular that of obtaining a satisfactory lubricating layer. Some of the techniques presented are considered to be suitable for polymer solutions only, e.g. Metzner (32, 83) and Williams (21).

The converging flow models are further detailed in Table 1 in terms of the form of the constitutive equations used to describe the shear and extensional behaviour, whether the geometries are axisymmetric (cylindrical) or planar (slit), and their application. A limited number of models incorporate elasticity. Binding (36), Gleissle (43) and Humphries et al (31) allow elasticity terms that are independent of any viscosity functions. The effect of elasticity is discussed in greater detail in Section 3.3. Further specific comments related to each model are also presented.

3.3. CONTRIBUTION OF ELASTICITY TO ENTRANCE PRESSURE DROPS

The nature of the fluid's properties that results in high entrance pressure drop values for non-Newtonian fluids was previously the subject of considerable debate. Two schools of thought existed: one considered that the pressure drop was due to elasticity effects, and the other considered that it was due to the fluid's resistance to elongational flow, characterised by the material's extensional viscosity.

Philippoff and Gaskins (44) interpreted the large entrance pressure drop (Bagley end correction) obtained for non-Newtonian viscoelastic fluids as being due to recoverable shear effects. This approach of accounting for the large entrance pressure drop in terms of elastic behaviour was subsequently adopted by La Nieve and Bogue (45), Boles et al (46), Okubo and Hori (35), Colbert and Ziegel (47) and Gleissle (43) for modelling die entry flow.

However, in more recent years the experimental and theoretical evidence, the later obtained through numerical modelling, indicates that the entrance pressure drop is dominated by the extensional flow behaviour of the melt rather than its elastic behaviour (35, 48-53). However, the principle shortcoming of these studies is that they tended to use normal stress difference data measured in shear as a measure of the elastic behaviour and correlated them with entrance pressure drop. This is perhaps because shear flow measurements of normal stress difference can be made using rotational rheometers that are in much more widespread use. Characterisation of the extensional viscoelastic properties is significantly more difficult. The material's extensional elastic behaviour may instead be the more relevant property to correlate with entrance pressure drop data. In numerical modelling studies extensional viscoelastic properties may be taken into account but only in as much as predicted through the use of constitutive equations (the shear and extensional properties may not be separable) and based on shear flow data. Nevertheless the following evidence is

² The entry angle is defined as that subtended by the die entry profile and the centreline of the geometry.

presented to illustrate the effect of elasticity, albeit characterised in shear, on the entrance pressure drop.

Mitsoulis (52), through numerical simulation of entrance flows, concluded that the high entrance pressure drops are due to the extensional viscosity behaviour of polymer melts rather than their elastic behaviour. Normal stresses (as generated in shearing flow and a measure of the fluid's elasticity) actually reduced the magnitude of the entrance pressure drop whereas higher extensional viscosities increased it. This observation was repeated by Binding (36). Mitsoulis (52) commented that the shear generated normal stresses "pull" the melt into the capillary thus reducing the pressure drop. He suggested that strain thickening behaviour may account for high entrance pressure drop values and, conversely, strain thinning behaviour for low entrance pressure drop values.

On the basis of numerical modelling and experimental work, Choplin et al (53) commented that it was not possible to correlate the recoverable shear with entrance pressure drop: they reported that a solution that exhibited large normal stresses in steady shear flow had entrance pressure drops of the same magnitude as a Newtonian fluid. They suggested that a correlation of entrance pressure drop values with a combination of elastic and shear thinning parameters is more likely as such fluids tend to exhibit large stress overshoots when submitted to a sudden and large shear flow. White et al (54) suggested that the origin of vortices was not related to the elasticity based on shear flow properties but due to the transient extensional viscosity behaviour.

Kwag et al (55) reviewed the origin of the entrance pressure drop and presented further evidence to the effect that normal stresses contribute negatively to the entrance pressure drop. The effect was demonstrated to be significant for model materials for which the ratio of extensional to shear viscosity was small although more realistic experimental data for elasticity were used. The extensional viscosity values derived using converging flow analysis were reduced by 50% when elasticity effects were taken into account. Unfortunately, in using extensional viscosity data for real materials (obtained from Laun and Schuch (56)) the effect of elasticity was not explored. In this case the contribution to the entrance pressure drop due to extensional viscosity would be expected to be considerably more significant thus reducing the relative effect of elasticity.

Binding (17) indicated, through use of his converging flow model, that under conditions of severe shear thinning ($n < 1/3$) elasticity may contribute positively to the entrance pressure drop. He commented that in the case of a fluid exhibiting a shear viscosity independent of shear rate ($n=1$) and a first normal stress difference proportional to the square of the shear rate the effect of elasticity may be significant. Polymer melts typically have n values between $1/3$ and $2/3$. Cogswell (16) commented that the elastic deformation of polymers becomes saturated at relatively low levels of stress compared with those found in converging flows, with the consequence that the effect on pressure drop due to elasticity was small compared with the effect of shear and extensional viscous dissipation.

It therefore appears that elasticity, characterised using shearing flows, normally reduces the magnitude of the entrance pressure drop for polymer melts. However, the magnitude of the effect is not clearly known although evidence suggests that typically it will be small. The effect of elasticity generated in extensional flow on entrance pressure drop measurements remains even less well unexplored.

3.4. GENERAL ASSUMPTIONS TYPICALLY MADE IN USING CONVERGING FLOW MODELS

The important aspects of converging flow models, in addition to the predictions being of value, are that the models are relatively easy to use and the data required are easily obtained. In applying contraction flow methods and comparing the results with stretching methods there are a number of assumptions that are made and these are discussed below.

The extensional viscosity of a polymer melt is a function of both strain rate and strain. Polymer melts exhibit either unbounded stress growth with increasing strain, a maximum in the stress growth curve or an equilibrium value at high strains (4). However, converging flow methods characterise materials flowing in a region in which the total strain of deformation is constant and is determined by the contraction ratio. For example, the Hencky strain occurring in a contraction having a 19:1 barrel to die diameter is calculated to be ≈ 5.9 . The comparison of extensional viscosities determined from stretching methods with contraction flow data is thus complicated by the fact that contraction flow methods ignore the effect due to strain and the strain history: the assumption is normally made that extensional viscosity is independent of strain and thus steady uniaxial extensional viscosity values are determined. A review of stretching methods (4) indicated that where steady extensional values were obtained it was at strains above approximately 4. This is less than that obtained in converging flow measurements for the above contraction ratio. Thus it is expected that, at best, agreement can only be obtained when the material exhibits a steady extensional viscosity at values of strain lower than that occurring in the converging flow, and that the converging flow pressure drop is dominated by the region of flow in which the strain is greater than that necessary for steady extensional viscosity. Perhaps fortuitously, Binding and Walters (57) commented that the pressure drop in a contraction flow occurred over a relatively small region close to the entry of the die. Nevertheless, for a polymer melt exhibiting strain hardening it is not expected that good agreement will necessarily be obtained.

Walters (58) commented that converging flows are "non-controllable" in that neither the stress nor strain rate are kept constant in the measurement region. For converging flow methods, although the total strain may be constant the strain rate typically is not constant, except when using a lubricated hyperbolic profiled die, Kim (24). Furthermore Walters (58) commented that consideration must be given to the deformation prior to the material entering the measurement region as it may have a significant influence on results. This latter comment is particularly relevant, for example, to spin-line measurements where the material is strained prior to testing.

As mentioned above, Binding and Walters (57) reported that the pressure drop in a contraction flow occurred over a relatively small region close to the entry of the die. This indicates that the positioning of the pressure transducer, i.e. sufficiently far upstream of the die entrance so that accurate measurements of entrance pressure drop are obtained, is not too critical.

The entrance pressure drop determined using a Bagley plot is comprised of terms due to both the entry region and the exit region. Forsyth (59) commented that the exit pressure drop is usually of the order of one tenth of the entry pressure drop for polymer melts. However the exit pressure drop is difficult to determine accurately due to various experimental problems, for example due to viscous heating and pressure dependence effects. It is normally assumed that the exit pressure loss is negligible and thus the value of the pressure drop determined using the Bagley plot is used as the entrance pressure drop.

The use of "zero" length dies can introduce measurement errors as such dies are more prone to damage and wear and are also more difficult to manufacture and calibrate. There are further arguments concerning the effect of the die on the flow field. A "zero" length die may have a slightly different entry flow field to that of a finite length die due to the additional constraint imposed by the internal wall of the die. Thus extrapolation using two dies of finite length to zero length may yield slightly different entrance pressure drop values to that obtained using a "zero" length die.

It is normally assumed that the converging flow is isothermal and that viscosities are pressure independent. Cogswell (60) commented that the effects of these two factors tend to cancel each other out. The effect of viscous dissipation on pressure drop can be estimated using a work done principle. However the author is unaware of any data on the pressure dependence of extensional viscosity, though it could be assumed to be the same as that for shear viscosity for which limited data exist (61).

In formulating the relationship between the entrance pressure drop data and the rheological properties further assumptions are often made. The shear and extensional viscosity data are fitted using simple constitutive equations, normally power-law models (see Table 1). The issues related to elasticity of the melt were discussed in Section 3.3. Furthermore the velocity field in the converging flow region is assumed. These are obviously approximations of the behaviour of polymer melts that is in reality far more complex in both shear and extension (62).

Inertia is normally assumed to be negligible although formulation of some of the converging flow models (34) has taken it into account. For polymer melts the magnitude of the effect is considered to be negligible.

Some of the errors incurred in using a converging flow model to determine extensional viscosity data may, fortuitously, be cancelled if those data are then used to calculate the pressure drop in another flow geometry. It is recommended that the same converging flow model is used for both the analysis of entrance pressure drop data and for their application.

3.5. CONVERGING FLOW MODELS : COMPARISON OF COGSWELL, GIBSON AND BINDING MODELS.

The models due to Cogswell [22], Gibson [23] and Binding [34] are the most widely used models reported in the literature (see Table 2). In comparing the formulation of these models the following observations are made.

Both Gibson [23] and Binding [34] assumed fully independent power-law equations to describe the shear and extensional viscosities whereas Cogswell [22] assumed that the extensional viscosity was constant and the shear viscosity was defined using a power-law. To determine the shear component of flow each assumed a fully developed shear flow velocity profile for their analyses. However, Gibson [23] used a spherical coordinate system (as opposed to cylindrical coordinates) in order to describe the problem with the consequence that a different "fully developed" velocity profile to that of the other models was obtained (the centre line velocity was greater in the Gibson model). With the exception of the Binding model, average strain rates were adopted in the formulation of these converging flow models. Thus Gibson and Cogswell departed from the fully developed shear flow velocity profile assumption in this respect. Binding however calculated the local value of the strain rate from the assumed velocity profile.

To determine the relationship between the entrance pressure drop data and extensional viscosity, and also the position of the vortex boundary for freely converging flow, Cogswell and Binding employed the principle of minimum work. The model proposed by Gibson is for application to constrained entry flow only and therefore cannot be used to predict a free convergence entrance angle. However, comparison of data with the prediction of the model indicates that the model may be used with entry angles up to 90° . For further details of these models see the Appendices 1-3.

In Cogswell's original model (22) apparent values of shear rate and shear viscosity are used. Kwag (55) and Kenig (63) reformulated the Cogswell model to take into account the non-Newtonian velocity profile (Rabinowitsch correction). Kwag (55) demonstrated that for a material having a low ratio of extensional to shear viscosity the incorporation of the Rabinowitsch correction would reduce the ratio further. Bersted (37) also modified the Cogswell model by relaxing two of the assumptions made therein, specifically by using a rate dependent extensional viscosity (power-law) and a shear viscosity function with a variable shear rate dependent power-law index n . As a result of the changes made by Bersted (37) numerical solution of the revised model is required. Similarly Tremblay (38) modified the Cogswell model to incorporate both power-law extensional viscosity and Rabinowitsch-corrected shear viscosity behaviour, see Appendix 1.

3.6. COMPARISON OF PREDICTIONS OF CONVERGING FLOW MODELS WITH OTHER METHODS

Validation of the predictions of converging flow models for polymer melts is very limited, Table 2. Validation has been presented either by comparison of results with other test methods (37, 56, 57, 64-69), by intercomparison of converging flow models (38, 55, 70-72) or by comparison with the predictions of numerical modelling (55). The most significant comparisons are those due to Laun and Schuch (56) and Bersted (37) (the latter using Laun and Schuch's (56) experimental data) in which good agreement was obtained for polyolefins at high rates but was worse at low rates. In both cases the Cogswell model under-predicted values at low rates compared with tensile measurements. The agreement of Cogswell's model with tensile measurements was qualitatively poor for polystyrene although values were of the correct magnitude.

Hingman et al (66) demonstrated good agreement of predictions using the Cogswell model with tensile measurements for polypropylene albeit over a limited strain rate range.

The work by Kwag et al (55), in which the predictions of the Cogswell and Binding models were compared with numerical modelling, indicated that the agreement was generally good, with those of the Binding model being consistently better and lower than the Cogswell model.

Bersted (37) demonstrated that an improved fit to experimental data (56) could be obtained using the Cogswell model by assuming a strain rate dependent extensional viscosity and a shear viscosity model incorporating a shear rate dependent n (power-law index). The fit was improved for polyolefins predominantly at lower strain rates but had relatively little effect at higher strain rates or for polystyrene. Both changes resulted in an improvement in the predictions although the effect due to the change in the extensional viscosity function was perhaps marginally greater.

4. DISCUSSION AND CONCLUSIONS

Numerous converging flow models for determining extensional viscosities from entrance pressure drop data are presented in the literature (Table 1). The most widely used models, as evident in the literature (Table 2), are those due to Cogswell (22), Binding (34) and Gibson (23) with Cogswell's being the most prevalent. However, there is insufficient evidence in the literature to strongly support a particular choice of one of the models of either Cogswell, Binding or Gibson, or to estimate with any reasonable confidence the level of agreement that may be obtained between converging flow and stretching methods. Many of the other models presented in Table 1 have limitations that may make them undesirable to use either from a theoretical or practical point of view.

The validity and assumptions associated with the converging flow approach and the advantages and disadvantages of the various models have been discussed. The contribution to the entrance pressure drop due to elasticity, characterised using shear flow, can be predicted using a modified version of the Binding model. Although the magnitude of the effect has not been clearly quantified for polymer melts the available evidence indicates that it is likely to be small.

The difficulties of reliably validating these models were discussed for polymer solutions and are assumed to be the same for polymer melts, i.e. extensional viscosity is normally a function of strain rate and strain. Thus, since different test methods tend to have different strain-time histories, comparison of results between methods is difficult. Nevertheless, validation of the Cogswell model against other methods is presented (Table 2) and indicated that good agreement can be obtained between stretching and converging flow methods. However, this was not always the case and the availability of such validation in the literature is extremely limited.

Better agreement was obtained for polyolefins at low strain rates when the Cogswell model was modified to incorporate a power-law extensional viscosity and a rate dependent power-law index n for shear viscosity (37). The latter describes more accurately the shear behaviour at low rates compared with a power-law model. However, these modifications had little effect on predictions at high strain rates. Furthermore, the formulation of the model by Bersted (37) requires numerical solution.

Despite the problems with, and limitations of converging flow methods the technique has considerable benefits as a materials characterisation method:

- it can provide valuable information on the extensional flow behaviour of melts,
- it is simple to use, and
- if capillary extrusion rheometry is to be used to determine shear flow behaviour then entrance pressure drop data and thus extensional viscosity data can be obtained at minimal additional cost.

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Table 1: Contraction flow models, Page 1 of 5.

Ref.	Author	Year	Solution or melt	Cylindrical or planar geometry	Entry geometry	Shear Functions	Extensional Functions	Materials, test conditions and comments
33	Balakrishnan et al	1976	solutions only	cylindrical	90°	not applicable	not assumed	Natural convergence angle required for solution. Shear forces ignored.
37	Bersted	1993	melt	cylindrical	90°	$\eta = \eta_o / (1 + a\dot{\gamma}^{2/3})$	$\lambda \propto \dot{\epsilon}^m$ where m is also a function of strain rate	LDPE, HDPE, PS. Development of Cogswell model. Iterative solver required. Converging flow model results in reasonable agreement with Laun and Schuch (56) - better for HDPE and LDPE than PS. Improvement due to use of rate dependent extensional viscosity c.f. Cogswell model.
34	Binding	1988	solution	both	90°	$\eta = \eta_o \dot{\gamma}^{n-1}$	$\lambda = \lambda_o \dot{\epsilon}^{m-1}$	Restricted and limited validation based on Newtonian fluid results. Predicts free convergence profile based on energy balance. Funnel flow with free convergence boundary.
36, (also in 17)	Binding	1991	solution	cylindrical	90°	$\eta = \eta_o \dot{\gamma}^{n-1}$ $N_1 = c \dot{\gamma}^{p+1}$ where N_1 is the first normal stress difference	$\lambda = \lambda_o \dot{\epsilon}^{m-1}$	Indicates the effects of extensional viscosity and elasticity are opposite: elasticity decreases Couette correction. Development of Binding (34). Effect of elasticity on pressure drop, not on profile. Both funnel flow and quasi-radial flow entry conditions (vortex and vortex-free flows) though latter not suited for determining extensional viscosity.
25	Binding et al	1989	solution	planar	hyperbolic profile: constant strain rate, lubricated flow	$\eta = \eta_o \dot{\gamma}^{n-1}$	$\lambda = \lambda_o \dot{\epsilon}^{m-1}$	Polyacrylamide solutions. Typically poor agreement with spin line measurements. Indication that flow was not in fact constrained but was freely converging within profiled boundary. Better agreement obtained when constrained flow occurred. Energy balance to converging flow rheometer analysis.

Table 1: Contraction flow models, Page 2 of 5.

Ref.	Author	Year	Solution or melt	Cylindrical or planar geometry	Entry geometry	Shear Functions	Extensional Functions	Materials, test conditions and comments
22	Cogswell	1972	melt	both	90° or conical profile	$\eta = \eta_o \dot{\gamma}^{n-1}$ (formulation with Rabinowitsch correction presented by Gibson (18))	$\lambda = \text{constant}$	LDPE, PP, Acrylic. No comparison of extensional viscosity values. Force balance. Predicts profile in free convergence.
26	Everage et al	1978	melt	cylindrical	lubricated, constant strain rate profile	not applicable	not assumed	PS. Shear forces ignored.
23 (also in 18)	Gibson	1989	melt	cylindrical	conical profile, though 90° results reported	$\eta = \eta_o \dot{\gamma}^{n-1}$	$\lambda = \lambda_o \dot{\epsilon}^{m-1}$	Nylon 66 (50%wt gf); PP (30%wt gf); unsaturated polyester bulk moulding compound (BMC) (14%wt gf). Can use 90° entry half-angle. Spherical co-ordinate system.
19	Gibson et al, Part 1	1985	melt	cylindrical	conical profile	$\eta = \eta_o \dot{\gamma}^{n-1}$	$\lambda = \lambda_o \dot{\epsilon}^{n-1}$	Unsaturated polyester bulk moulding compound (BMC). Modified Cogswell model presented using power-law viscosities. Same rate dependence for shear and extensional viscosity. Cylindrical coordinates used. Validity restricted to entry angle values less than natural convergence angle. Correction for viscous temperature rise in determining entrance pressure drop.
20	Gibson et al, Part 2	1985	melt	cylindrical	conical profile	$\eta = \eta_o \dot{\gamma}^{n-1}$	$\lambda = \lambda_o \dot{\epsilon}^{n-1}$	BMC. Model same as ref. 19 except that spherical co-ordinate system used. Best fit of model for entry angle < 30°. Fit to pressure drop profile over full range of entry angles up to 90° was poor.

Table 1: Contraction flow models, Page 3 of 5.

Ref.	Author	Year	Solution or melt	Cylindrical or planar geometry	Entry geometry	Shear Functions	Extensional Functions	Materials, test conditions and comments
43	Gleissle	1988	melt	cylindrical	geometry taken into account using experimentally determined geometry factor	normal stress difference related to shear viscosity	not applicable	LDPE. Relates entrance pressure drop to the first normal stress difference, given a geometry factor determined experimentally.
39	Huang et al	1981	melt	both	lubricated profiled	not assumed	not assumed	LDPE. Relationship between stresses and pressure drop. No strain rates and thus extensional viscosities calculated.
31	Humphries et al	1985	melt	both	conical profiled, converging - parallel-diverging flow geometry	$\eta = \eta_o \dot{\gamma}^{n-1}$ $G = G(\tau)$	$\lambda = \lambda(\dot{\epsilon})$	LDPE, HDPE, PS, PP. Requires accurate pressure measurement to determine elastic component. Analysis assumes reversible elastic component and single extensional viscosity function for both converging and diverging flows.
24	Kim et al	1994	melt	planar	hyperbolic-constant strain rate profiled converging die	not applicable	Newtonian or power law fluid models	LDPE skin/PP core. No determination of elongational viscosity presented - reported that research into this still ongoing. Lubricated flow yielding essentially pure elongational flow. Numerical solution required. Also $\eta_{core} > 100 \times \eta_{lubricant}$.
32	Metzner et al	1969	solutions only	cylindrical	90°	not applicable	Maxwell model	Jet thrust technique applicable to solutions only. Shearing forces ignored.
83	Metzner et al	1970	solutions only	cylindrical	90°	not applicable	not assumed	Jet thrust technique applicable to solutions only. Shearing forces ignored.

Table 1: Contraction flow models, Page 4 of 5.

Ref.	Author	Year	Solution or melt	Cylindrical or planar geometry	Entry geometry	Shear Functions	Extensional Functions	Materials, test conditions and comments
35	Okubo et al	1981	melt	cylindrical	90°	not applicable	simple viscoelastic model	HDPE. Simple viscoelastic model (two Voigt elements in series). Shear forces neglected. Converging flow angle restricted to less than 60°. Normal force calculated from entry flow behaviour.
27	Shaw	1975	melt	cylindrical	lubricated, conical profile	not applicable	simple viscoelastic model	LDPE, HDPE. Perfectly lubricated assumption. Maximum extensional stresses determined from which pressure drop was then determined numerically. Extensional viscosities not presented.
28	Snelling et al	1960	melt	cylindrical	lubricated conical profile	$\tau_{\max} = C\dot{\gamma}_{\max}^n + \eta\dot{\gamma}_{\max}^m$	$\sigma_{\theta\theta} - \sigma_{rr} = 2\tau_{\max}$	PTFE. Entrance pressure drop values calculated from materials parameters rather than reverse done.
38	Tremblay	1989	melt	cylindrical	90°	$\eta = \eta_0 \dot{\gamma}^{n-1}$	$\lambda = \lambda_0 \dot{\epsilon}^{m-1}$	LDPE, LLDPE, LDPE/LLDPE blend. Two models presented: Development of Cogswell (22) model to incorporate power-law behaviour of extensional viscosity and non-Newtonian shear flow velocity profile. Approximate sink flow analysis based on energy dissipated in viscous flow. Factor in model needs to be determined from free convergence entry angle, and then used to calculate extensional viscosity parameters. Application best suited therefore to transparent systems only.

Table 1: Contraction flow models, Page 5 of 5.

Ref.	Author	Year	Solution or melt	Cylindrical or planar geometry	Entry geometry	Shear Functions	Extensional Functions	Materials, test conditions and comments
21	Williams et al	1985	solution	planar	constant strain rate profiled	not applicable	not assumed	Lubricated flow producing planar orthogonal stagnation flow. Pressure drop and geometry used to determine extensional viscosity. Experimentally difficult to operate. Comprehensive list of conditions that are required to be met was presented.
29	Williamson et al	1984	melt	cylindrical	conical profile	contribution due to shear flow pressure drop considered negligible.	$\lambda = \text{constant}$	Unsaturated polyester dough moulding compound (DMC). Radial flow. Validity restricted to entry angle values less than natural convergence angle. Recommended $\alpha < 30^\circ$.
30	Zahorski	1992	solution	planar	profiled	Incompressible, simple fluid: Newtonian and slightly non-Newtonian fluids.	Incompressible, simple fluid: Newtonian and slightly non-Newtonian fluids.	Lubricated die converging flow rheometer analysis for slightly non-Newtonian fluids or numerical solutions. Method as for Williams et al (21) but includes correction for shear effects in the flow. Constitutive form needs to be assumed.

TABLE 2: Usage of converging flow models and comparison of predictions with other results and experimental techniques for polymer melts: Page 1 of 5

Ref.	Author	Year	Converging flow model	Materials	Comparison with other techniques	Conclusions / comments
80	Beery et al	1992	Gibson (23)	PC, LCP, PC/LCP blend.	No comparison with other methods.	$\lambda/\eta = 50 - 200$ for PC, 100-300 for blend, 300 -400 for LCP. Use of different entry angles had no effect on entrance pressure drop - possibly material forming own convergent region within entry cone geometries
37	Bersted	1993	Cogswell (22) Bersted's (37) development of Cogswell (22)	LDPE, HDPE, PS.	Laun and Schuch (56) tensile test results.	Converging flow results were in reasonable agreement with Laun and Schuch (56) data - better for HDPE and LDPE than PS. Improvement in agreement due to use of rate dependent extensional viscosity and more suitable shear viscosity function c.f. Cogswell model.
57	Binding	1988	Binding (34)	polymer solutions	Comparison with spin-line measurements	Converging flow analysis was a useful method for estimating extensional viscosity of shear thinning polymer solutions. However for constant viscosity polymer solutions converging flow analysis yielded poor agreement (factor of x20 difference in values).
73	Binding	1991	Binding (34)	long glass fibre filled PP, PP.	Comparison with theory due to Goddard (74) assessing the contribution of rigid particles (fibres) to the extensional flow behaviour of the material.	A factor of approx 2.5 lower than predicted values (Goddard theory) due to the considerable difficulties in quantifying the fibre aspect ratio. Taking into account fibre degradation can improve level of agreement (also see Rides (75)). λ/η up to 1000, $\lambda/\eta = 8$ for unfilled PP ($\lambda/\eta = \text{extensional viscosity} / \text{shear viscosity}$).
64	Cogswell	1972	Cogswell (22)	LDPE, HDPE, LDPE/HDPE blend, PP	Cam loading, constant stress extensional rheometer (77).	Questionable indication of agreement for LDPE only as extrapolation of results from constant stress tensile rheometer to converging flow analysis stress range was required.
65	Covas et al	1990	Cogswell (22)	HDPE, LDPE, LLDPE, PS, PVC.	Converging flow analysis data compared with extensional stretching flow data (Rutherford extensional rheometer). Calculated vortex angles from the analysis are compared with values from flow visualisation studies.	No overlap of strain rates to compare directly extensional stretching results with Cogswell model predictions. Indicated high sensitivity of predictions to shear flow and entrance pressure drop input parameters. Converging flow analysis predictions were approximately one decade higher than tensile measurements (inferred by extrapolating results) except for LDPE for which extrapolation would indicate reasonable agreement. Confirmed qualitative validity of Cogswell model as relative extensional behaviours determined by converging flow analysis and stretching techniques were consistent.

TABLE 2: Usage of converging flow models and comparison of predictions with other results and experimental techniques for polymer melts: Page 2 of 5

Ref.	Author	Year	Converging flow model	Materials	Comparison with other techniques	Conclusions / comments
72	Groves et al (private communication)	1997	Cogswell (22) Gibson (18) but with shear and extensional rate dependence parameters the same.	LDPE, HDPE	Comparison of Binding with Cogswell model predictions.	Authors comment that the differences between the predictions of the models are small if their limitations, specifically relating to the entry flow / geometry angle, are observed. Cogswell free convergence model can be used for entry angles less than 90° provided the flow is freely converging within the geometry. This implies that the effect of the vortex is small.
66	Hingmann et al	1994	Cogswell (22)	PP	Comparison with uniaxial stretching rheometer.	Good agreement of extensional viscosity values in the region of the strain rate overlap. Authors limited converging flow analysis to higher entrance pressure drop values to minimise errors in analysis.
63	Kenig	1991	Cogswell (22) with Rabinowitsch correction for non-Newtonian shear flow velocity profile.	PC, LCP.	No comparison with other models.	PC and LCP exhibit power-law behaviour in extension.
70	Kiang et al	1992	Binding (34) Cogswell (22)	PET	Converging flow analysis models compared.	Extensional viscosity has same strain rate (thinning) dependency in both models, but Trouton ratios using Cogswell model approx. 70% higher than Binding model (in range $\approx 7 - 10$ for the Cogswell model). Trouton ratio relatively unchanged with extrusion rate, leading to absence of vortex growth. Results consistent with flow visualisation experiment.
55	Kwag et al	1991	Cogswell (22) Binding (34)	LDPE, HDPE.	Theoretical comparison of extensional values used in finite element calculations of entrance pressure drop with values determined using converging flow analysis from the calculated entrance pressure drops, using Cogswell and Binding models.	Extensional viscosity values determined using the Cogswell model were consistently higher than values determined using Binding method, except for when the Trouton ratio was set at a value of 3. Authors comment that the Cogswell model was not very accurate, but still acceptable for stretch rate above 10/s. Authors were unaware of any better method and commented that the Binding model is more complicated to apply and does not significantly improve on the Cogswell model. Improvement would be made by incorporating the contribution due to normal stresses. (The author considers that, on the basis of the data presented, the Binding model yielded consistently better fit to the data inputted into the finite element model.
78	La Mantia et al	1982	Cogswell (22)	HDPE/ LDPE blends	No comparison with other methods.	Evaluation of blends for blown films. Correlation of relative behaviour of blends with $3\eta_0$ values.

TABLE 2: Usage of converging flow models and comparison of predictions with other results and experimental techniques for polymer melts: Page 3 of 5

Ref.	Author	Year	Converging flow model	Materials	Comparison with other techniques	Conclusions / comments
56	Laun et al	1989	Cogswell (22)	HDPE, LDPE, PS.	Converging flow analysis compared with other types of elongational rheometry measurements: Rheotens fibre spinning and steady-state viscosities obtained using constant strain rate and constant stress uniaxial extensional rheometers.	Qualitative agreement of converging flow analysis results with Rheotens extensional viscosities although converging flow analysis results were lower than Rheotens values. Correlation with fibre drawability made. Converging flow data were in good agreement with constant strain and constant stress data for LDPE and an HDPE, although agreement was worse at low strain rates ($< 1 \text{ s}^{-1}$). For another HDPE and PS the agreement was worse, but values were of the correct magnitude. Authors commented that no general rule on the range of agreement can be given and they cautioned against drawing further conclusion from converging flow data.
71	Padmanabhan et al	1994	Binding (34) (planar and uniaxial) Cogswell (22) (planar and uniaxial) Gibson (23) (uniaxial only)	LDPE	Converging flow analysis models compared.	In-line measurement using slit (planar flow) and capillary (cylindrical flow) dies. Cogswell and Binding models showed similar trends but Cogswell values consistently higher by 40% in the planar case but slightly lower by $\approx 20\%$ in the uniaxial case. In the uniaxial case Gibson's analysis showed completely different trend and magnitude. Various possible explanations of these observation presented. However, an abrupt contraction was used for which the Gibson model was perhaps not best suited. Cogswell model recommended because of simpler equations.
40	Pendse et al	1995	Kim (50): lubricated converging flow technique.	PP	No comparison with other methods.	Uses commercially available rheometer. $\lambda/\eta \approx 30$.
41	Pendse et al	1996	Kim (50): lubricated converging flow technique.	PP, sPS, nylon-66.	No comparison with other methods.	Uses commercially available rheometer.

TABLE 2: Usage of converging flow models and comparison of predictions with other results and experimental techniques for polymer melts: Page 4 of 5

Ref.	Author	Year	Converging flow model	Materials	Comparison with other techniques	Conclusions / comments
42	Pendse et al	1996	Kim (50): lubricated skin-core profiled converging flow approach.	PP, sPS, Nylon-66.	Predicted pure elongational flow in converging flow confirmed by tracer particles and image analysis.	Planar and uniaxial measurements reported. Used two extruders for the planar flow and a capillary extrusion rheometer for uniaxial flow. In the planar case (strain rates up to 0.5 s ⁻¹) Trouton ratios of up to ≈2000 for PP presented. In uniaxial case (strain rates up to 500 s ⁻¹) Trouton ratios were up to 100. Conditions for developing pure elongational flow are flow rate ratio of core/skin layers~10, and viscosity ratio at least 100.
81	Pickles et al	1996	Gibson (23)	LCP, glass fibre filled LCP.	No direct measurement comparison made. Speculative assessment using Goddard theory with little conclusive outcome.	Used Gibson model over restricted shear rate ranges of data due to their non-linearity. $\lambda/\eta \approx 30$.
76	Sherbelis et al	1996	Binding (34)	PP	Based on use of extensional data to predict flow in a nozzle.	Stated that "Binding's model has been shown to be a significant improvement [on Cogswell's]", but with no supporting evidence as to why.
67	Shroff et al	1977	Cogswell (22)	HDPE, LDPE PP, PS	Comparison with isothermal melt spinning. Also comparison with vortex angle at die entry.	Quantitative agreement. For the worst case reported η_e was within ≈40%. Similar vortex angle. Flow activation energies were within ≈15% for extension and shear for LDPE, PS and PP: similar to observation by Münstedt (79) on PS melts. Authors observed that there was a correlation between the ratio of the entrance pressure to shear stress ($\Delta P/\tau$) and the vortex entry angle θ .

TABLE 2: Usage of converging flow models and comparison of predictions with other results and experimental techniques for polymer melts: Page 5 of 5

Ref.	Author	Year	Converging flow model	Materials	Comparison with other techniques	Conclusions / comments
38	Tremblay	1989	Cogswell (22): model revised to incorporate power-law behaviour of extensional viscosity and non-Newtonian shear flow velocity profile. Binding (34) Tremblay (38): approximate sink flow analysis.	LDPE, LLDPE, LDPE / LLDPE blend.	Binding and Cogswell models compared with approximate sink flow analysis (ASF). Predictions of all three models were compared with predictions of Larson model: non-linear viscoelastic model incorporating strain hardening based on dynamic data and given values for the strain hardening parameter.	Binding and ASF analyses gave same (to within 10%) elongational viscosity for LLDPE. Cogswell model yielded higher extensional viscosity by $\approx 50\%$ and lower strain rates than the other two models. Entry angle were reasonably predicted using the Binding and Cogswell models, but not by ASF, when the entrance angle was small and at low strain rates (large vortices). ASF analysis gave better agreement at large entry angles for LLDPE. Difficulty lies in determination of value of the factor F of the ASF model. For LDPE, ASF gave a strain rate thinning behaviour yet Cogswell and Binding models yielded strain rate thickening behaviour - authors comment that the former behaviour is more in agreement with other literature although conclusive evidence not presented. Reliability of comparison with Larson model questionable as model needs to be extrapolated beyond measured data. Extensional viscosity has same dependence on deformation rate for both modified Cogswell and Binding models. All three converging flow models predicted vortex growth if $m > n$.
82	Tremblay	1992	Tremblay (58): approximate sink flow analysis.	Various LDPE / LLDPE blends.	No comparison with other methods.	Observed a correlation of the difference in shear and extensional rate dependence parameters (m-n) with concentration of LDPE in the blend.
68	Turek et al	1993	Cogswell (22)	semicrystalline LCP, amorphous LCP.	Isothermal melt spinning	Reasonable quantitative agreement for semicrystalline LCP (maximum x2 discrepancy), although spin-line measurements indicated strain hardening and converging flow analysis indicated strain thinning. However the level of agreement for amorphous LCP (x50 discrepancy presented) cannot be assessed due to the testing temperatures differing by 10 °C.
69	Tzogankis et al	1989	Cogswell (22)	PPs (at varying levels of free-radical-induced degradation).	Compared with observations of fibre spinning.	Authors commented that PPs that had elongational viscosity almost independent of strain rate, indicated using the Cogswell model, could be drawn more successfully.

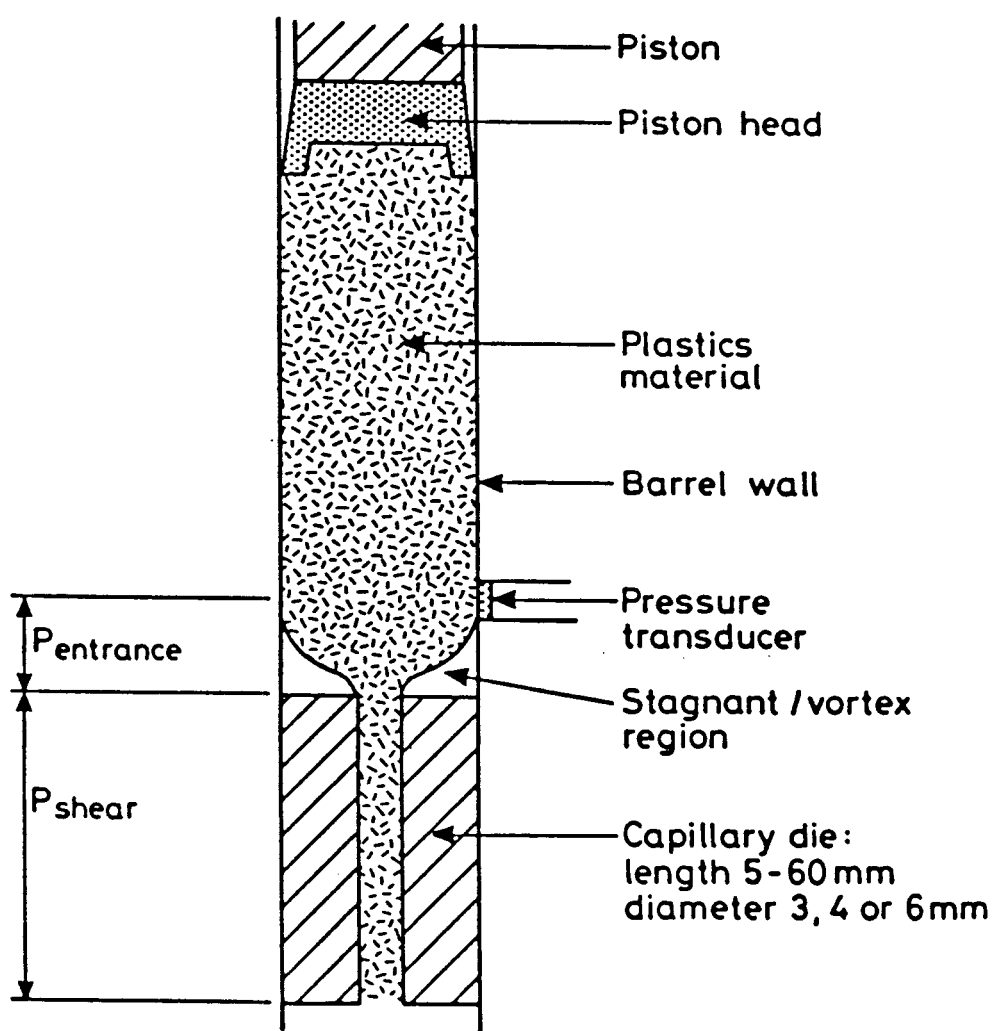


Figure 1: Capillary extrusion rheometer showing the entrance pressure drop and shear flow pressure drop components of the flow and the vortex (secondary flow) region.

APPENDICES: DESCRIPTION OF SELECT CONVERGING FLOW MODELS

For reasons of consistency the notation used in these appendices is not necessarily the same as that used in the original references (22, 23 and 34).

APPENDIX 1: COGSWELL MODEL

The converging flow model developed by Cogswell (22) has two versions; one is applicable to the analysis of free converging flow, the other to constricted converging flow. Free convergence occurs when the flow establishes its own primary-secondary flow boundary within the confines of the contraction region (primary refers to the main flow through the die and secondary refers to any vortex or stagnant region formed in the die entry region). Constricted converging flow occurs when no secondary flow region is formed, such as when using an entry cone of angle less than that of the naturally occurring primary-secondary flow boundary. Both cases are considered below.

In the converging flow model it was assumed that shear viscosity was described by the power-law behaviour

$$\eta = \eta_o' \dot{\gamma}^{n-1} \quad (\text{A1.1})$$

and extensional viscosity λ was independent of strain rate

$$\lambda = \lambda_o' \quad (\text{A1.2})$$

where η_o' and λ_o' were constant for a given temperature. The analysis considered that the converging flow region was comprised of a series of short truncated conical elements of decreasing diameter. The velocity profile of the flow in each element was assumed to be the same as that for fully developed shear flow. The shear flow pressure drop for the element was determined by integration over the length of the tapered element. The extensional flow pressure drop was based on the average extension rate over the length of the element, and as such was independent of the assumed velocity profile. The shear flow pressure drop ΔP_s and the extensional flow pressure drop ΔP_E for an element of entry radius R_i , exit radius R_o ($R_o < R_i$) and converging half-angle α are given by

$$\Delta P_s = 2 \frac{(\eta_{app}|_{r=R_o})}{3n \tan \alpha} \left[\frac{4Q}{\pi R_o^3} \right] \left[1 - \left(\frac{R_o}{R_i} \right)^{3n} \right] \quad (\text{A1.3})$$

and

$$\Delta P_E = \frac{\lambda \tan \alpha}{3} \left[\frac{4Q}{\pi R_o^3} \right] \left[1 - \left(\frac{R_o}{R_i} \right)^3 \right] \quad (\text{A1.4})$$

where $\eta_{app}|_{r=R_o}$ denotes the apparent shear viscosity evaluated for the apparent wall shear-rate for flow rate Q in a channel of radius R_o . The total pressure drop ΔP_o for the element was calculated, assuming additivity, as the sum of the shear and extensional flow pressure drops.

Thus

$$\Delta P_o = \Delta P_s + \Delta P_E \quad (A1.5)$$

For constricted converging flow the total pressure drop P_o in an entry region of geometry defined by an entry radius R_i , exit radius R_o and converging half-angle α is also given by Equations A1.3-A1.5 (omitting the increment notation). Given the shear viscosity power-law coefficients the extensional viscosity λ can be determined using

$$\begin{aligned} P_o - P_s &= P_E \\ &= \frac{\lambda \tan \alpha}{3} \left[\frac{4Q}{\pi R_o^3} \right] \left[1 - \left(\frac{R_o}{R_i} \right)^3 \right] \end{aligned} \quad (A1.6)$$

where P_s is calculated using Equation A1.3 and P_o is the experimentally measured entrance pressure drop.

The maximum average strain rate $\dot{\epsilon}_{av}$ is related to the apparent wall shear-rate at the exit ($r = R_o$) by

$$\dot{\epsilon}_{av} = \frac{\tan \alpha}{2} \left(\frac{4Q}{\pi R_o^3} \right) \quad (A1.7)$$

These equations have been used in the analysis of entrance pressure drop data obtained using constricted converging flow geometries.

For freely converging flow the total pressure drop in the element ΔP_o (Equation A1.5) was minimised with respect to the converging angle α of the element. The total pressure drop over the entire converging flow region was then given by the sum of the minimised pressure drops for all elements, taking the limit of this summation as the length of the individual elements tended to zero. The minimised total pressure drop P_o for free convergence is given by the expression

$$P_o = \frac{4\sqrt{2}}{3(n+1)} \left(\frac{4Q}{\pi R_o^3} \right) \left[\left(\eta_{app} \times \lambda \right) \Big|_{r=R_o} \right]^{\frac{1}{2}} \quad (A1.8)$$

the die entry half-angle α_o ($r=R_o$) by

$$\tan \alpha_o = \left[\left(\frac{2\eta_{app}}{\lambda} \right) \Big|_{r=R_o} \right]^{1/2} \quad (A1.9)$$

and the primary-secondary boundary profile by

$$h = \frac{\sqrt{2} r}{(3n-1)} \left[\left(\frac{r}{R_0} \right)^{\frac{3n-1}{2}} - 1 \right] \left[\left(\frac{\lambda}{\eta_{app}} \right) \Big|_{r=R_0} \right]^{\frac{1}{2}} \quad (A1.10)$$

where h is the upstream distance from the die entry plane and r the radial position. The average strain rate $\dot{\epsilon}_{av}$ at the die entrance ($r=R_0$) is given by

$$\dot{\epsilon}_{av} = \left(\frac{2Q}{\pi R_0^3} \right) \left[\left(\frac{2\eta_{app}}{\lambda} \right) \Big|_{r=R_0} \right]^{\frac{1}{2}} \quad (A1.11)$$

In deriving this model it was assumed that the extensional viscosity was independent of strain rate. However, if the extensional viscosity of the fluid is calculated for each flow rate then an estimate of the strain rate dependence of extensional viscosity can be obtained.

Tremblay (38) reformulated Cogswell's free convergence model to incorporate an independent power-law extensional viscosity function (eg. Equation A2.2) and shear viscosity corrected for non-Newtonian velocity profile (Rabinowitsch correction)*. This yielded

$$P_0 = \left\{ 4^{2m} \left(\frac{4}{3} \right)^{m-1} \left(\frac{32}{9} \right) \lambda_0 \eta_0^m \left(\frac{n}{3n+1} \right)^{2m} \right\}^{\frac{1}{1+m}} x^{\frac{\dot{\gamma}^{\frac{m(1+n)}{1+m}}}{1+n}} \quad (A1.12)$$

$$\text{where } \dot{\gamma} = \left[\frac{3n+1}{n} \right] \left[\frac{Q}{\pi R_0^3} \right] \quad (A1.13)$$

Equation A1.12 compares with equation A1.8 of the original Cogswell model.

Note: * the expression presented in Tremblay (38) is missing the brackets around the fraction $32/9$ and is thus incorrectly expressed therein.

APPENDIX 2. GIBSON MODEL

Gibson (23) developed a model for determining the extensional viscosity of fluids from constricted converging flow data only. However, he proposed that the model could be used for entrance pressure drop data obtained using contraction regions with entry half-angle α of up to 90° . The author commented that although the secondary flow region had a significant effect on the appearance of the flow field in the entry region, it had only a small effect on the entrance pressure drop. Thus no distinction between constricted and free convergence data was made. The model cannot be used to predict the free convergence primary-secondary flow boundary.

The converging flow model allows for both the shear and extensional viscosities to be described by independent power-law models:

$$\eta = \eta_o' \dot{\gamma}^{n-1} \quad (\text{A2.1})$$

$$\lambda = \lambda_o' \dot{\epsilon}^{m-1} \quad (\text{A2.2})$$

where λ_o' is a function of temperature, $\dot{\epsilon}$ is the strain rate and m is a constant. The analysis of the converging flow region was carried out by dividing the region into two parts A and B, Figure A2.1. In region A the pressure drop contributions due to both the shear and extensional flow of the fluid were considered. In region B only the pressure drop contribution due to extensional flow was considered. The total pressure drop was calculated as being the sum of the shear component $P_{A,S}$ and the extensional components $P_{A,E}$ and $P_{B,E}$. These pressure drop components are given by Equations A2.3, A2.4 and A2.6 respectively. The shear flow pressure drop was calculated on the basis of a fully developed shear flow velocity profile transformed onto a spherical boundary coordinate system. The strain rate used was an average for the section, Equation A2.5, and was therefore independent of the assumed velocity profile. The three components of pressure drop in the contraction region are:

$$P_{A,S} = \frac{2\eta_o' \left(\dot{\gamma}_{w,app}^n \Big|_{r=R_o} \right) \sin^{3n} \alpha}{3n \alpha^{(1+3n)}} \left(\frac{1+3n}{4n} \right)^n \left(1 - \left(\frac{R_o}{R_i} \right)^{3n} \right) \quad (\text{A2.3})$$

$$P_{A,E} = \frac{2\lambda_o'}{3n} (\dot{\epsilon}_1)^m \left(1 - \left(\frac{R_o}{R_i} \right)^{3m} \right) \quad (\text{A2.4})$$

where the average strain rate $\dot{\epsilon}_1$ is given by

$$\dot{\epsilon}_1 = \left(\dot{\gamma}_{w,app} \Big|_{r=R_o} \right) \frac{\sin \alpha (1 + \cos \alpha)}{4} \quad (\text{A2.5})$$

and

$$P_{B,E} = \frac{\lambda_o' \left(\dot{\gamma}_{w,app} \Big|_{r=R_o} \right)}{4^m} \phi(m, \alpha) \quad (\text{A2.6})$$

where

$$\dot{\gamma}_{w,app} \Big|_{r=R_o} = \frac{4Q}{\pi R_o^3} \quad (\text{A2.7})$$

and

$$\Phi(m, \alpha) = \int_0^\alpha (1 + \cos \beta)^{m-1} \sin^{m+1} \beta d\beta \quad (\text{A2.8})$$

$\Phi(m, \alpha)$ was solved by numerical iteration. The total entrance pressure drop P_o is given, assuming additivity, as

$$P_o = P_{A,S} + P_{A,E} + P_{B,E} \quad (\text{A2.9})$$

If this is rewritten in terms of the pressure drop due to extensional flow in regions A and B, then

$$P_o - P_{A,S} = P_{A,E} + P_{B,E} \quad (\text{A2.10})$$

Thus, substituting for $P_{A,E}$ and $P_{B,E}$ using Equations A2.4 and A2.6,

$$P_o - P_{A,S} = \lambda_o' \left(\dot{\gamma}_{w,app}^m \Big|_{r=R_o} \right) \left[\left(\frac{2}{3m} \right) \left(\frac{\sin \alpha (1 + \cos \alpha)}{4} \right)^m \left(1 - \left(\frac{R_o}{R_i} \right)^{3m} \right) + \phi(m, \alpha) / 4^m \right] \quad (\text{A2.11})$$

P_o is the experimentally measured entrance pressure drop and $P_{A,S}$ is calculated using Equation A2.3 and appropriate values for the shear viscosity power-law coefficients. A plot of $\log(P_o - P_{A,S})$ versus $\log(\dot{\gamma}_{w,app} \Big|_{r=R_o})$ has a gradient m and intercept value Y_{int} where Y_{int} is given by

$$Y_{int} = \lambda_o' \left[\left(\frac{2}{3m} \right) \left(\frac{\sin \alpha (1 + \cos \alpha)}{4} \right)^m \left(1 - \left(\frac{R_o}{R_i} \right)^{3m} \right) + \frac{\phi(m, \alpha)}{4^m} \right] \quad (\text{A2.12})$$

Thus λ_o' can be determined.

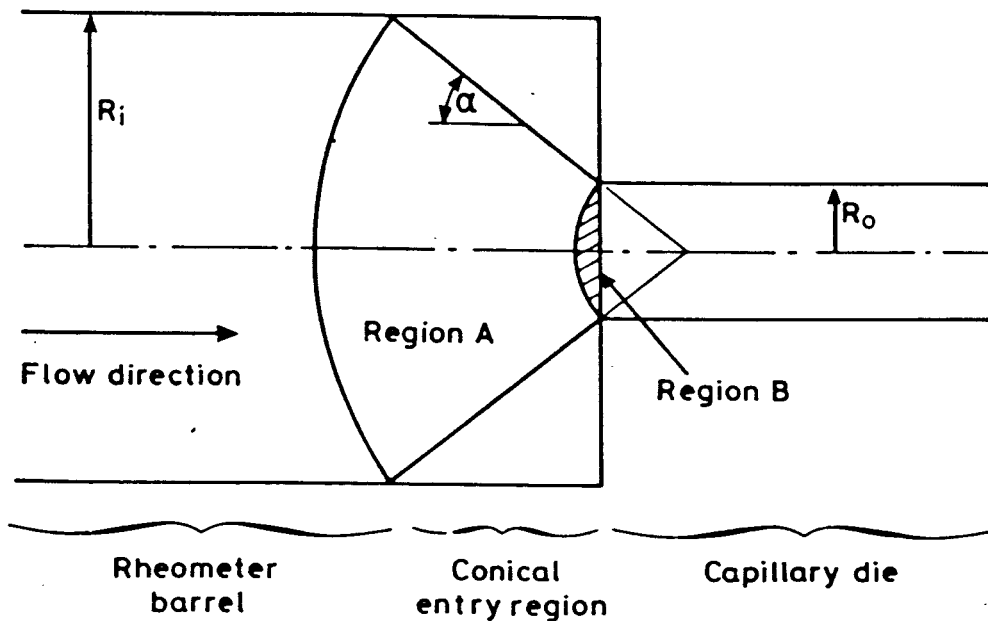


Figure A2.1 Entry geometry definition for the Gibson model (23).

APPENDIX 3. BINDING MODEL

The approach used by Binding (34) was to consider the minimisation of the rate of change of energy occurring in the converging flow of a fluid. This change in energy results from the process of viscous dissipation and also from a change in kinetic energy of the fluid. The model applies to free convergence flow only. Independent power-law models identical to those used by Gibson (23) were used to describe the shear and extensional viscosities, Equations A2.1 and A2.2. A fully developed power-law shear flow velocity profile was assumed. Local shear and extensional deformation rates $\dot{\gamma}$ and $\dot{\epsilon}$ were derived using this velocity profile and are given by

$$\dot{\epsilon} = \left(\frac{3n+1}{n+1} \right) \frac{Q}{\pi R^3} \left(\left(\frac{3n+1}{n} \right) \left(\frac{r}{R} \right)^{1+1/n} - 2 \right) \frac{dR}{dz} \quad (\text{A3.1})$$

and

$$\dot{\gamma} = - \left(\frac{3n+1}{n} \right) \frac{Q}{\pi R^3} \left(\frac{r}{R} \right)^{1/n} \quad (\text{A3.2})$$

where z and r are defined by a cylindrical coordinate system, Figure A3.1, and R defines the primary-secondary flow boundary (dR/dZ is negative in value). It was noted that $\dot{\epsilon}$ is positive at $r = 0$, and negative at $r = R$. The calculated negative strain rates in the converging flow model arise as a consequence of the assumed velocity profile: they are not expected to occur in practice.

Expressions for the shear and extensional viscous dissipations in a conical element of the converging flow field were derived using Equations A3.1 and A3.2. The expression for the rate of change of kinetic energy of the fluid, Equation A3.5, was also derived from consideration of the velocity field. However, this contribution is considered to be minimal for polymer melts and can usually be ignored. Thus the rate of change of energy of the fluid flowing in the converging section was determined as the sum of the rate of shear and extensional viscous dissipations, this being obtained by integration of the dissipation terms over the flow volume. This was then minimised with respect to the shape of the primary-secondary flow boundary to yield the flow field relating to minimum energy dissipation. The entrance pressure drop P_0 in the converging section was related to the rate of change of energy of the system $\dot{E}$ by

$$P = \frac{\dot{E}}{Q} \quad (\text{A3.3})$$

By substituting for the minimised rate of change of energy $\dot{E}$ the entrance pressure drop was given by

$$P_o = \frac{2\dot{\eta}_o (1+m)^2}{3m^2 (1+n)^2} \left(\frac{\dot{\lambda}_o m(3n+1) n^m I_{nm}}{\dot{\eta}_o} \right)^{1/(1+m)} \dot{\gamma}_o^{m(n+1)/(1+m)}$$

$$\times \left(1 - \alpha^{3m(1+n)/(1+m)} \right) + \frac{K\dot{E}}{Q} \quad (A3.4)$$

where the rate of change of kinetic energy $K\dot{E}$ was given by

$$K\dot{E} = \frac{3\rho Q^3 (3n+1)^2 (1-\alpha^4)}{2(2n+1)(5n+3)\pi^2 R_o^4} \quad (A3.5)$$

where ρ is the fluid density. Also

$$\dot{\gamma}_o = \left(\frac{3n+1}{4n} \right) \left(\frac{4Q}{\pi R_o^3} \right) \quad (A3.6)$$

$$\alpha = \left(\frac{R_o}{R_i} \right) \quad (A3.7)$$

$$I_{nm} = \int_0^1 \left[\text{abs} \left(2 - \left(\frac{3n+1}{n} \right) \varphi^{1+1/n} \right) \right]^{m+1} \varphi d\varphi. \quad (A3.8)$$

The optimised primary-secondary flow boundary relating to minimum energy dissipation was defined by

$$\frac{dR}{dz} = - (n+1) \left(\frac{k}{\ell m(3n+1) n^n I_{nm}} \right)^{1/(m+1)} \left(\frac{(3n+1)Q}{\pi R^3} \right)^{\frac{n-m}{m+1}} \quad (A3.9)$$

or alternatively

$$L_v = \frac{(1+m)}{2(n+1)(2m-3n-1)} \left(\frac{\dot{\lambda}_o (3n+1) m n^m I_{nm}}{\dot{\eta}_o} \right)^{1/(1+m)} \times \dot{\gamma}_o^{(m-n)/(1+m)} \times \left(1 - \alpha^{(2m-3n-1)/(1+m)} \right) \times \alpha \quad (A3.10)$$

From Equations A3.1 and A3.9 the strain rate everywhere in the converging flow region can be determined. The gradient G of a plot of $\log (P_o)$ versus $\log (\dot{\gamma}_o)$ was given (using Equation A3.4) by

$$G = m(n+1)/(1+m) \quad (\text{A3.11})$$

Thus, given the value of the shear viscosity exponent n , m can be determined. The integral I_{nm} , Equation A3.8, can then be evaluated numerically. The value of the intercept Y_{int} of the $\log(P_o)$ axis of the plot described above is given (using Equation A3.4) by

$$Y_{\text{int}} = \log \left[\frac{2\eta_o'(1+m)^2}{3m^2(1+n)^2} \left(\frac{\lambda_o' m(3n+1)n^m I_{nm}}{\eta_o'} \right)^{1/(1+m)} \right. \\ \left. \times (1 - \alpha^{3m(1+n)/(1+m)}) \right] \quad (\text{A3.12})$$

Thus λ_o' can then be calculated.

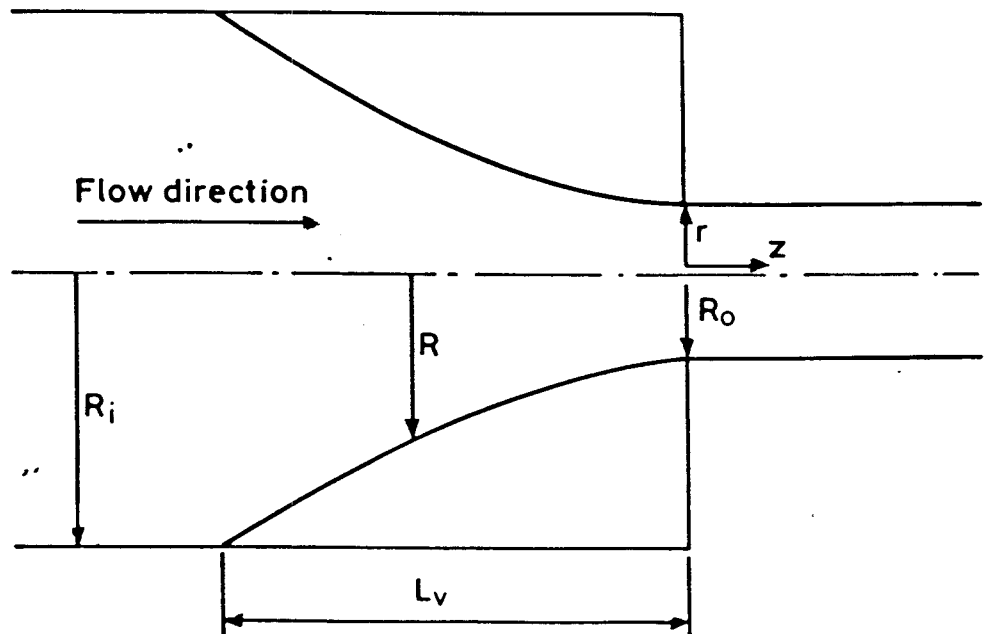


Figure A3.1 Entry geometry definition for the Binding model (34).