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capillary rheometer entrance pressure drop measurements
using a numerical simulation of contraction flow**

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ABSTRACT

Various experimental techniques are available for characterising the extensional flow behaviour of fluids, for example uniaxial stretching, squeeze flow and converging flow. In practice each technique is applicable to a relatively restricted range of fluids, and also to limited and often difficult to control test conditions. High viscosity filled polymer melts pose particular measurement problems. The preferred technique for this group of materials is the use of converging flow analysis to determine extensional viscosity from contraction flow pressure drop data which are readily obtained from capillary extrusion rheometry. Currently, only simplified analytical methods are available for interpreting these data.

A numerical model based on an augmented spectral method has been developed to simulate the flow in contraction regions of inelastic fluids which exhibit a high extensional viscosity to shear viscosity ratio. The model has been used to evaluate the use of an existing analytical model for determining extensional viscosity values. The evaluation demonstrates that the extent of agreement between the analytical and the numerical approach depended on the fluid behaviour, however the agreement was in general poor. The use of the spectral numerical contraction flow model, in place of analytical models, for determining extensional viscosity data from capillary entrance pressure drop data is shown to be an improvement on the analytical technique examined giving good agreement with experiment in the case of a DMC slurry. However, limitations in the technique have been identified for use with fibre-filled materials. Recommendations to extend the approach by using other constitutive equations in the contraction flow model and so enable application of the technique to a wider range of fluids have been made.

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FIGURES

1. INTRODUCTION

Extensional flow of plastics occurs in many industrial processes, for example in extrusion and injection moulding. To be able to model these processes reliably, and thus assist the plastics process designer through the provision of reliable CAD systems, the ability to characterise the extensional flow behaviour of the materials to be processed is essential.

Various experimental techniques are available for characterising the extensional flow behaviour of fluids, for example uniaxial stretching, biaxial stretching, film blowing, fibre spinning, open syphon, triple jet, squeeze flow and converging flow. In practice each technique is applicable to a relatively restricted range of fluids, and to a limited and often difficult to control range of test conditions.

High viscosity filled polymer melts pose particular measurement problems. The preferred technique for this group of materials is the use of converging flow analysis to determine extensional viscosity from contraction flow pressure drop data, which are readily obtained from capillary extrusion rheometer measurements by using capillary dies of different length, Figure 1, and extrapolating the pressure drop results to obtain that for a zero length die in a Bagley plot. A typical plot as in Figure 2 (1) shows the large entrance pressure drop values (at $L/R = 0$) obtained for a glass-fibre filled unsaturated polyester dough moulding compound (DMC) containing 6mm length fibres. Entrance pressure drop values as a function of apparent shear rate for various batches of this material are presented in Figure 3. The apparent shear rate is given by $4Q/\pi R^3$ where Q is the volume flow rate and R is the radius of the capillary die. Currently, only simplified analytical methods are available for interpreting such data (2-20). The predictions of the models due to Cogswell (2), Gibson (3), and Binding (4) have been compared in detail by Rides elsewhere (1): the extensional viscosity curves obtained using these analytical models for a glass-fibre filled unsaturated polyester dough moulding compound are presented in Figure 4. For this material there is a difference factor of approximately two in extensional viscosity values calculated from entrance pressure drop data using the Cogswell and Gibson models. In the original formulation of these analytical models approximate assumptions about flow conditions were necessarily made so that tractable solutions could be obtained. The aim of this work has been to improve on such techniques for determining extensional viscosity values of polymeric materials.

2. CONTRACTION FLOW NUMERICAL MODEL

A numerical model based on an augmented spectral method has been developed to simulate the flow in axisymmetric contraction regions of inelastic fluids which exhibit a high extensional viscosity to shear viscosity ratio (high Trouton ratio). The constitutive model used in the spectral code was

$$\mathbf{T} = 2\eta(\dot{\Gamma}, \dot{E})\mathbf{d} \quad [1]$$

where $\mathbf{T}$ is the extra-stress tensor, $\mathbf{d}$ is the rate of deformation tensor, and the function $\eta(\dot{\Gamma}, \dot{E})$ is defined by

$$\eta(\dot{\Gamma}, \dot{E}) = K[1+(\lambda_1\dot{\Gamma})^2]^{(n-1)/2} \times [1+(\lambda_2\dot{E})^2]^{(m-1)/2} \quad [2]$$

with $\dot{\Gamma} = 2(\text{II})^{1/2}$ and $\dot{E} = 3 \text{ III}/\text{II}$ where $\text{II} = (\frac{1}{2} \text{tr } \mathbf{d}^2)$, $\text{III} = \det \mathbf{d}$ and K , λ_1 , n , λ_2 , m are constants for the fluid.

In pure shear equation [2] yields

$$\eta = K[1+(\lambda_1\dot{\gamma})^2]^{(n-1)/2} \quad [3]$$

and in pure extension

$$\eta = 3K[1+3(\lambda_1\dot{\epsilon})^2]^{(n-1)/2} \times [1+(\lambda_2\dot{\epsilon})^2]^{(m-1)/2} \quad [4]$$

where $\dot{\gamma}$ is the shear rate and $\dot{\epsilon}$ the strain rate.

This form of constitutive equation has been chosen as it can be used to describe a wide range of rheological behaviours and it allows the extensional and shear viscosity values to be independently controlled thus permitting high Trouton ratios to be obtained by appropriate selection of the values for the parameters. The numerical model is described in detail by Davies et al. (5). The two geometries considered and the boundary conditions used are presented in Figures 5 and 6 and are referred to as Geometry 1 and Geometry 2 respectively. The model for Geometry 1 was developed because it was the geometry predominantly used in the analytical models referred to in section 1 and was thus the most suitable for evaluation of those models.

3. RESULTS AND DISCUSSION

3.1 EVALUATION OF THE BINDING ANALYTICAL CONTRACTION FLOW MODEL

The Geometry 1 analysis has been employed to evaluate the use of the Binding model (4) for determining extensional viscosity values as follows. The spectral analysis numerical code with the constitutive equation defined by [1] and [2] was used to calculate the entrance pressure drop versus flow rate relationship for flow of various fluids in this axisymmetric contraction region. The pressure along the centre-line of the geometry as a function of distance z from the capillary entrance / barrel exit is plotted for two flow rates in Figure 7. The initially increasing and then decreasing gradient of the pressure profile was considered to be the result of the imposed boundary condition at the exit, namely that the velocity profile at the barrel exit was equivalent to that for a fully developed shear flow. This increase in gradient was not observed in equivalent plots for the Geometry 2 model for which the fully developed shear flow exit condition was specified at the exit of the capillary die, rather than at the exit of the barrel. The numerical entrance pressure drop values were obtained by linearly extrapolating the upper part of the curve to the pressure axis ($z = 0$) as demonstrated in Figure 7. As such it represents the additional pressure drop contribution due to the contraction flow in excess of that for fully developed shear flow in the barrel and is the value required for analysis using the Binding model. The graph shows that the majority of the extensional pressure drop occurred over a relatively short distance from the contraction exit plane.

The Binding model (4) was then used to determine extensional viscosity values from the numerically calculated entrance pressure drop versus flow rate data and the shear viscosity parameters K , λ_1 and n . The Binding model is a free-convergence contraction flow model with the shear and extensional viscosities taken as independent power-law functions of shear rate and strain rate respectively, ie a linear relationship of $\log$ (stress) versus $\log$ (shear or strain rate), with the consequence that the derived relationship of $\log$ (entrance pressure drop) versus $\log$ (flow rate) is also linear. The values determined using the Binding model were then compared with extensional viscosity values calculated using equation [4] and the spectral model input parameters (K , λ_1 , n , λ_2 and m). A range of materials parameters was selected for this exercise to represent the range of behaviours exhibited by polymers. Results indicated that for the cases studied the agreement was generally poor: the best agreement obtained is shown in Figures 8 and 9. An example of the poor agreement that was obtained is presented in Figure 10. The extensional viscosity values obtained in using the Binding model were particularly sensitive to the selection of the flow rate range of the numerically

calculated entrance pressure drop data: in interpreting pressure drop data using the Binding model a flow rate range must be selected over which the relationship of log (entrance pressure drop) versus log (flow rate) is taken to be linear. The discontinuous extensional viscosity curves obtained using the Binding model in Figures 8 and 9 are based on values obtained using different flow rate ranges. Additional difficulties associated with the use of the analytical converging flow models have been discussed in (4).

A comparison of the pressure drop versus flow rate behaviour for Geometries 1 and 2 is presented in Figure 11. The use of Geometry 2 resulted in lower predicted pressure drop values than for Geometry 1. This is to be expected as in using Geometry 1 a more severe constraint on the flow was employed, namely that of a fully developed shear flow velocity profile on the exit of the contraction region, rather than on the exit of the capillary die as was the case for Geometry 2. This observation shows that the assumption made in the Binding model (and also in other models referred to in Section 1) that the flow at the exit of the contraction region is fully developed is in error and can have a marked effect on extensional viscosity values thus determined.

3.2 NUMERICAL APPROACH TO DETERMINING EXTENSIONAL VISCOSITY VALUES FROM ENTRANCE PRESSURE DROP DATA

As a consequence of the findings presented above the contraction flow spectral model for Geometry 2 has been used to circumvent the need for the assumptions made in the approximate analytical models. An algorithm was created for determining extensional viscosity values that was based on a data array of selected values for the shear viscosity parameters (K , λ_1 and n), the additional extensional viscosity parameters (λ_2 and m), and calculated spectral model entrance pressure drop versus flow rate data. The algorithm was used to obtain a least-squares best fit to experimental entrance pressure drop versus flow rate data and thus values for the extensional viscosity parameters λ_2 and m were derived, given the shear viscosity parameters.

This approach is demonstrated for an unsaturated polyester slurry (equivalent to the DMC but without glass-fibres), Figure 12. This shows that the Trouton ratio is approximately three at the low rates but increases as the rate increases. The value of three is that expected for a Newtonian fluid. To check the procedure and the accuracy of the numerical array, the determined extensional viscosity parameters and the shear viscosity parameters (Figure 12) have been specifically used in the contraction flow model to calculate the pressure drop

versus flow rate behaviour in the contraction region. The calculated data are compared with the original experimental results in Figure 13. The agreement was generally good, in this case being within 5%, confirming that the approach followed and the numerical array used was reliable. However the agreement was not so good when deriving values of the parameters outside the range used to create the numerical array, as is to be anticipated when extrapolating rheological behaviour.

A more severe test was then carried out for the model. The entrance pressure drop data for the unsaturated polyester DMC materials are considerably higher than for the equivalent slurries (1). Use of the analytical contraction flow models suggests Trouton ratios of the order of 30 - 70 for these materials. The experimental entrance pressure drop versus flow rate data for a 19 mm diameter barrel to 4 mm diameter die contraction were used to derive extensional viscosity parameters. These values, along with shear viscosity values, were then used to calculate the expected entrance pressure drop values for the 19:4 contraction; and also to predict those for a 19 mm barrel to 6 mm die contraction, the experimental results for which were known. Figure 14 shows that for the 19:4 contraction there is relatively poor agreement of the experimental and predicted values compared with the slurry. Also the predicted pressures for the 19:6 contraction ratio are also in poor agreement with experimental values, Figure 15.

It would appear that the chosen model is unable to simulate the rapid change in gradient of the pressure curves at low flow rates: in contraction flow the material exhibits a "yield-like" behaviour at low flow rates (Figure 14 and 15), although this was not observed to be the case in shear flow. This behaviour is not too surprising if one considers the nature of the material, which contains a high concentration of 6 mm long glass fibre bundles. In capillary rheometry testing of this material 'log-jamming' occurred, particularly at low flow rates (1), with fibres preventing flow into the capillary die. The result is a temporary or permanent blockage of the die causing rapid increase in extrusion pressure. It is considered that a modification of the constitutive equation would improve the agreement obtained for this difficult DMC material. By increasing the power to which the term $\lambda_2 \dot{\epsilon}$ is raised in the constitutive equation [2] the shape of the pressure curve could be modified to give a better fit to the experimental data. To produce a more flexible option would require, for example, an equation of the form

$$\eta = 3K[1+3(\lambda_1 \dot{\epsilon})^2]^{(n-1)/2} \times [1+|\lambda_2 \dot{\epsilon}|^p]^{(m-1)/p} \quad [5]$$

4. CONCLUSIONS

A contraction flow numerical model using augmented spectral methods has been developed and has been used to evaluate the Binding analytical contraction flow model (4). This work has demonstrated that the latter model was not satisfactory for determining extensional viscosity values in the cases examined. A particular problem encountered in using the Binding model was the high sensitivity of the calculated extensional viscosity parameters to the selection of the flow rate range of pressure drop values. The Binding model formulation is based on fluids that exhibit a power-law behaviour in shear and extension with the consequence that the log (entrance pressure drop) versus log (flow rate) behaviour is itself linear. Thus complications arise in attempting to use the analytical model to interpret data that do not reasonably fit this behaviour.

To provide an alternative contraction flow analysis, the spectral model has been used to establish an algorithm that can be used to determine extensional viscosity parameters, given entrance pressure drop versus flow rate and shear viscosity data for the material. The contraction flow model is flexible in that the chosen constitutive model can represent a wide range of shear and extensional viscosity behaviours. The numerical array developed to determine the extensional viscosity parameters has been applied to results for an unsaturated polyester slurry, and has been shown to give a fit to experimental entrance pressure drop values to within 5%. Trouton ratios of the order of three were obtained at low flow rates. However the determination of the extensional viscosity of a long glass-fibre filled unsaturated polyester dough moulding compound proved less satisfactory. In checking the calculations of extensional viscosity parameters by re-calculating the entrance pressure drop data, poor agreement was obtained with the experimental results. This was due to the high sensitivity of the measured pressure drop data to flow rate at low flow rates - approaching a "yield-like" behaviour. Predictions of the pressure drop for a smaller contraction ratio also gave, not unexpectedly, a poor agreement with experimental results. To improve upon the capability of the numerical model to yield extensional viscosity values for these fibre-filled materials an improved form for the constitutive equation has been proposed.

5. ACKNOWLEDGMENT

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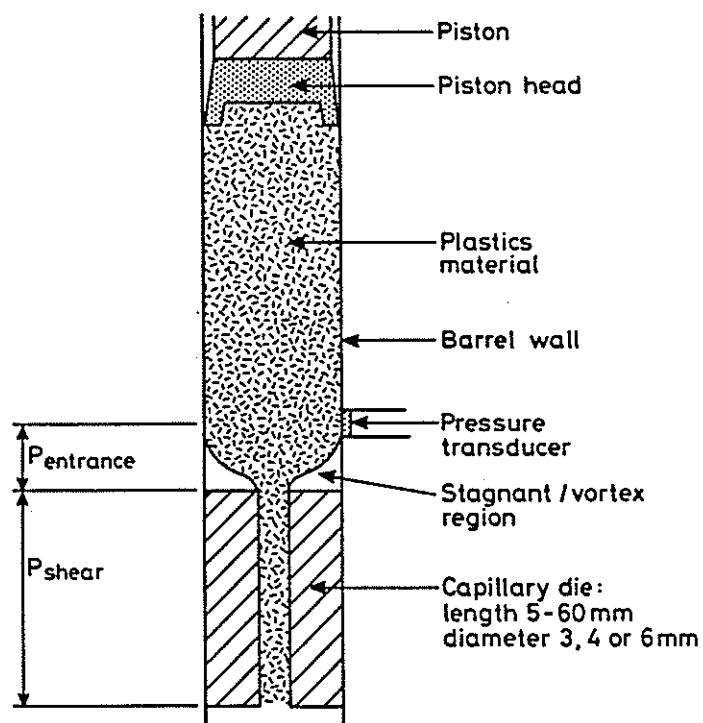


Fig 1. Contraction flow in a capillary rheometer

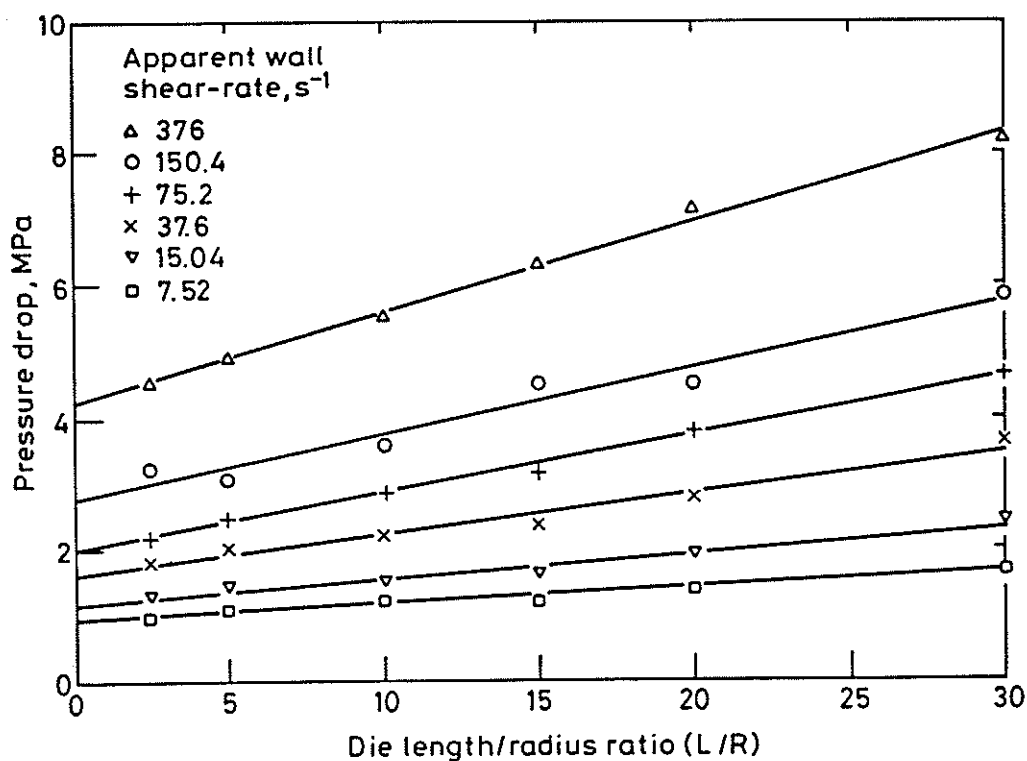


Fig 2. Bagley plot for a polyester DMC (L7049III) at 50 °C, using a 90° half-angle entry geometry.

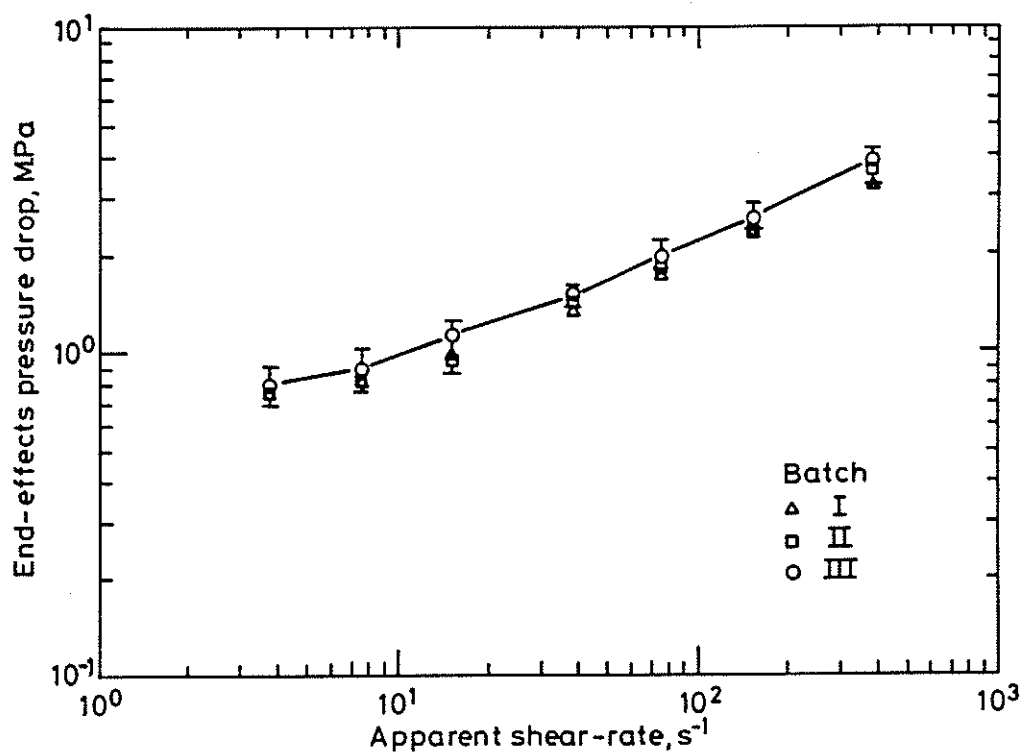


Fig 3. Comparison of mean entrance pressure drop measurements for DMC L7049 batches I, II and III at 50 °C. Scatter bands indicate the range of values obtained for batch III (eight tests at each shear rate).

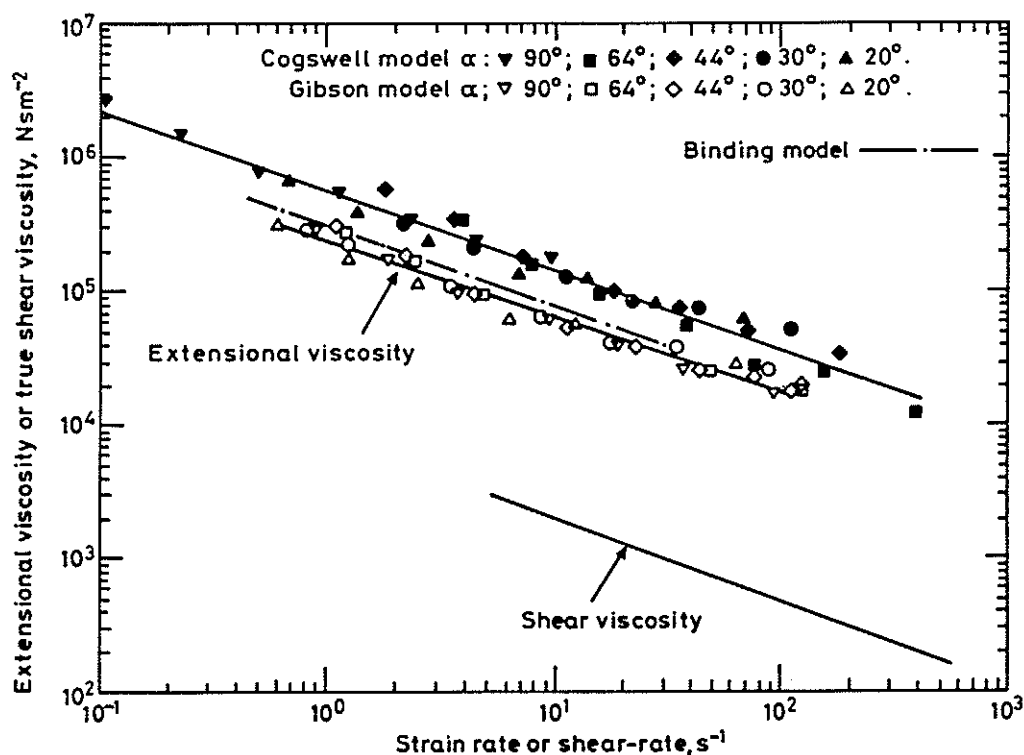


Fig 4. Comparison of extensional viscosity values obtained from entrance pressure drop measurements using the Cogswell, Gibson and Binding models, where α is the contraction entry half-angle.

Boundary conditions: no slip; isothermal;
fully developed shear flow
at entry and exit planes

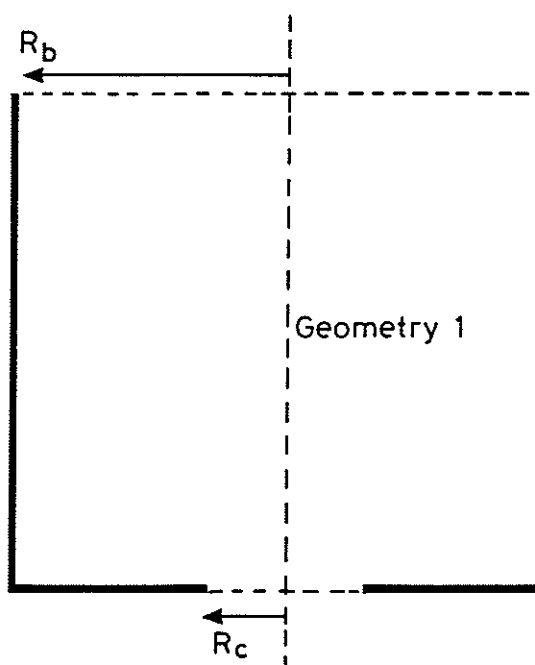


Fig 5. Geometry 1 used for numerical modelling of contraction flow for validation of analytical models.

Boundary conditions: no slip; isothermal;
fully developed shear flow
at entry and exit planes

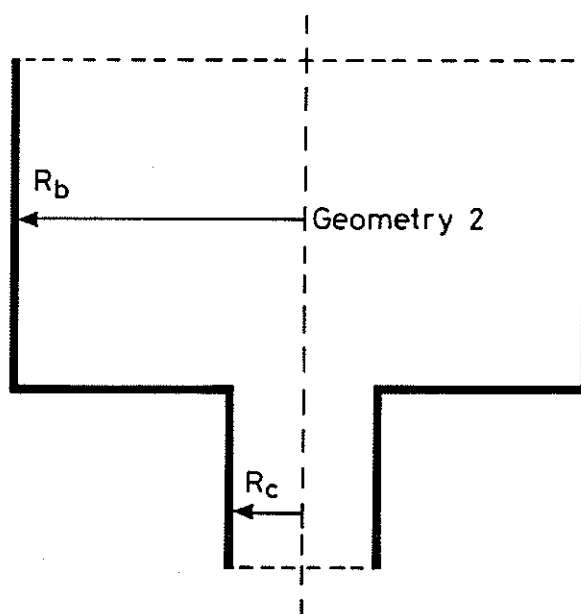


Fig 6. Geometry 2 used for numerical modelling of the full contraction flow geometry (rheometer barrel and capillary die).

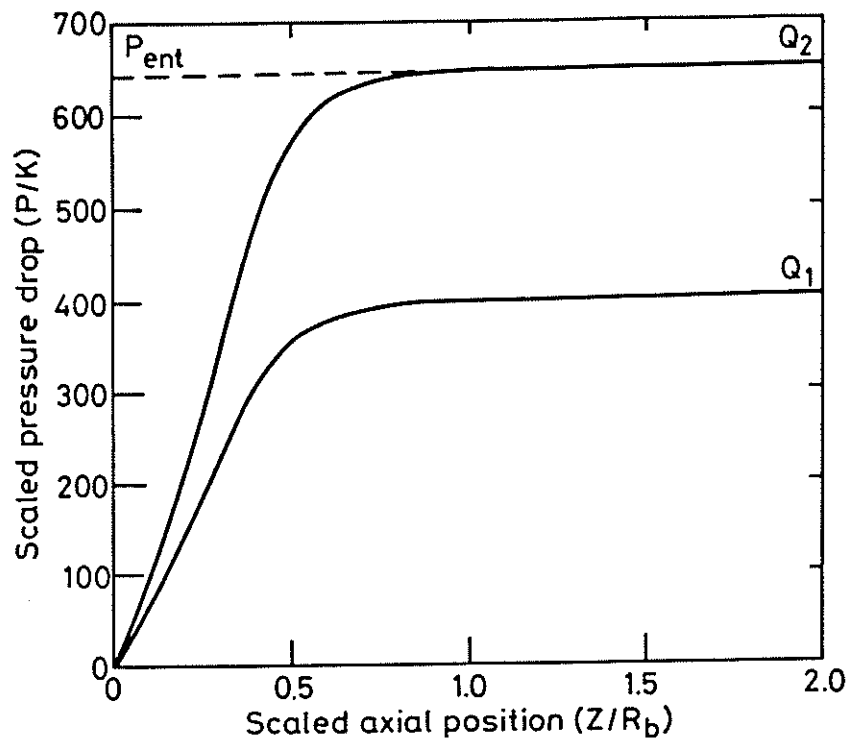


Fig 7. Pressure distribution upstream of the capillary die entrance, along the axis of symmetry. Test case: $n=1$, $\lambda_2 = 0.04$ s, $m = 1.5$, 4:1 diameter contraction ratio, $Q_1/(R_b^3) = 2.2$, $Q_2/(R_b^3) = 3.14$.

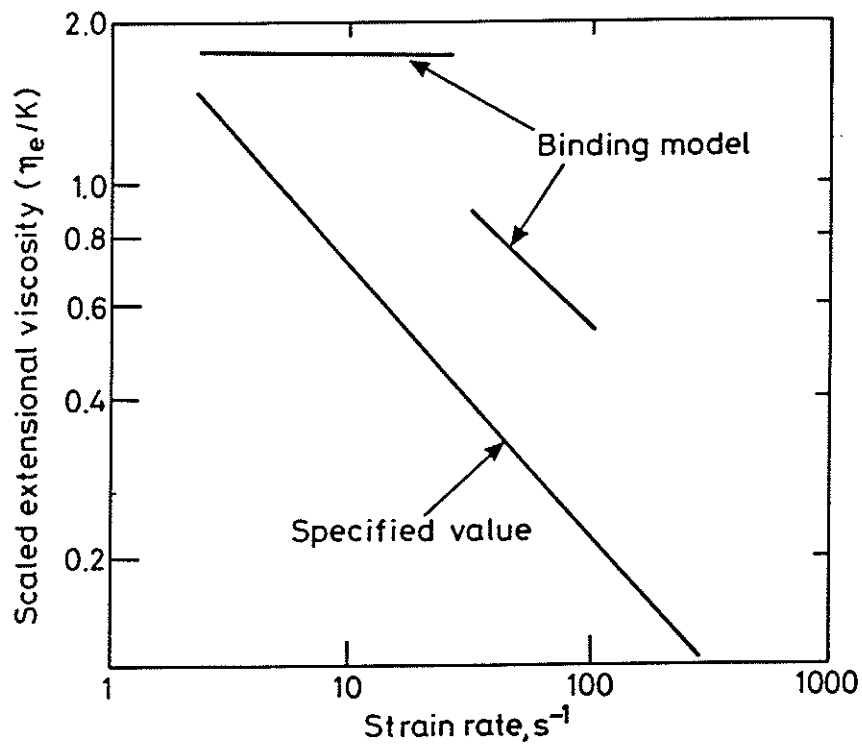


Fig 8. Comparison of specified spectral method extensional viscosity values with values determined using the Binding model over two shear rate ranges. Test case: $\lambda_1 = 1$ s, $n = 0.5$, $m = 1$, 4:1 diameter contraction ratio.

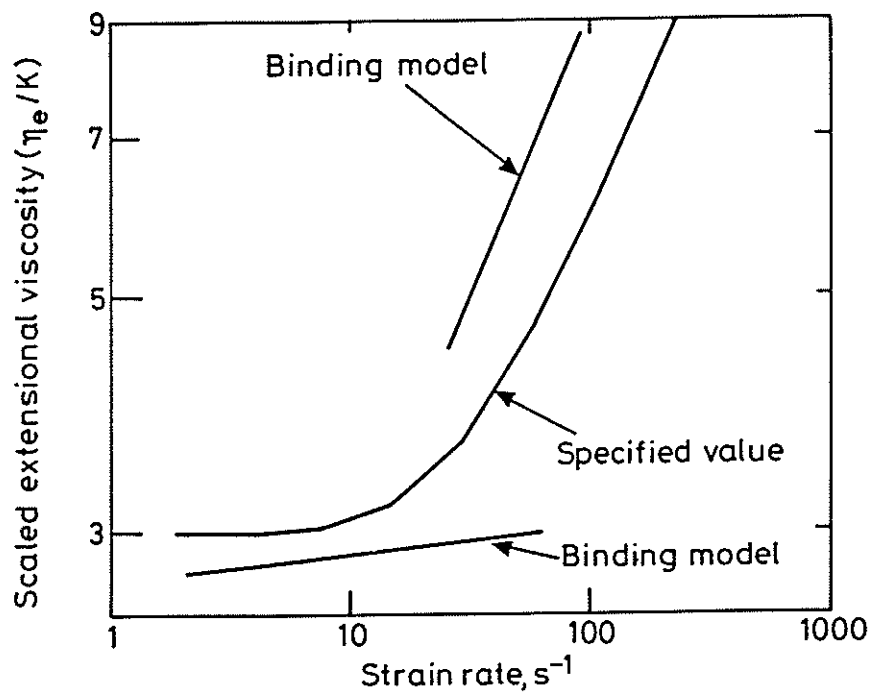


Fig 9. Comparison of specified spectral model extensional viscosity values with values determined using the Binding model over two shear rate ranges.
Test case: $n=1$, $\lambda_2 = 0.04$ s, $m = 1.5$, 4:1 diameter contraction ratio.

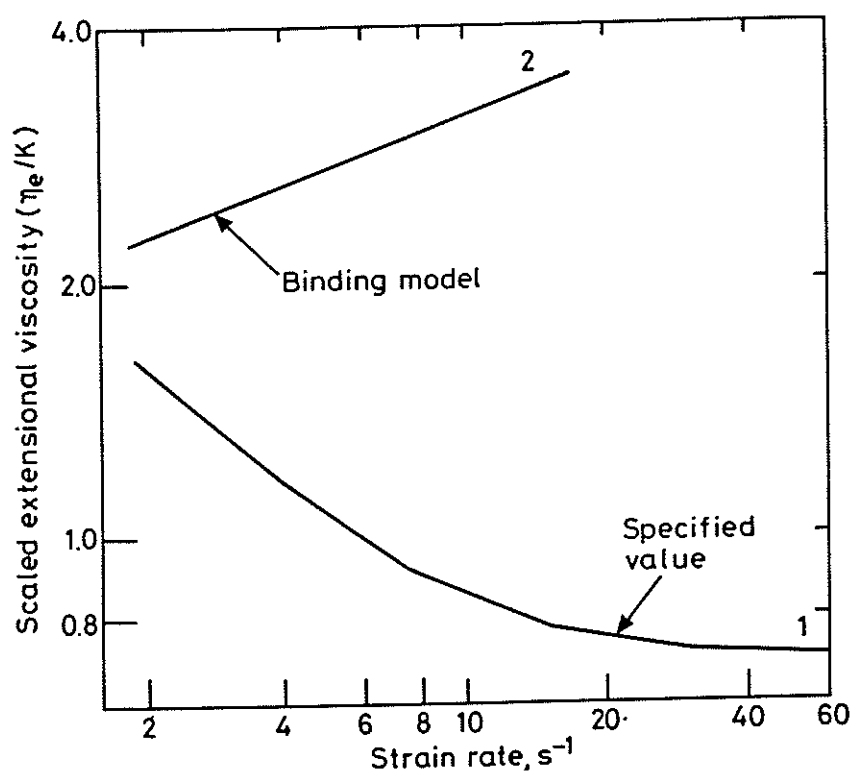


Fig 10. Comparison of specified spectral method extensional viscosity values with values determined using the Binding model.
Test case: $\lambda_1 = 1$ s, $n = 0.5$, $\lambda_2 = 0.1$ s, $m = 1.5$, 4:1 diameter contraction ratio.

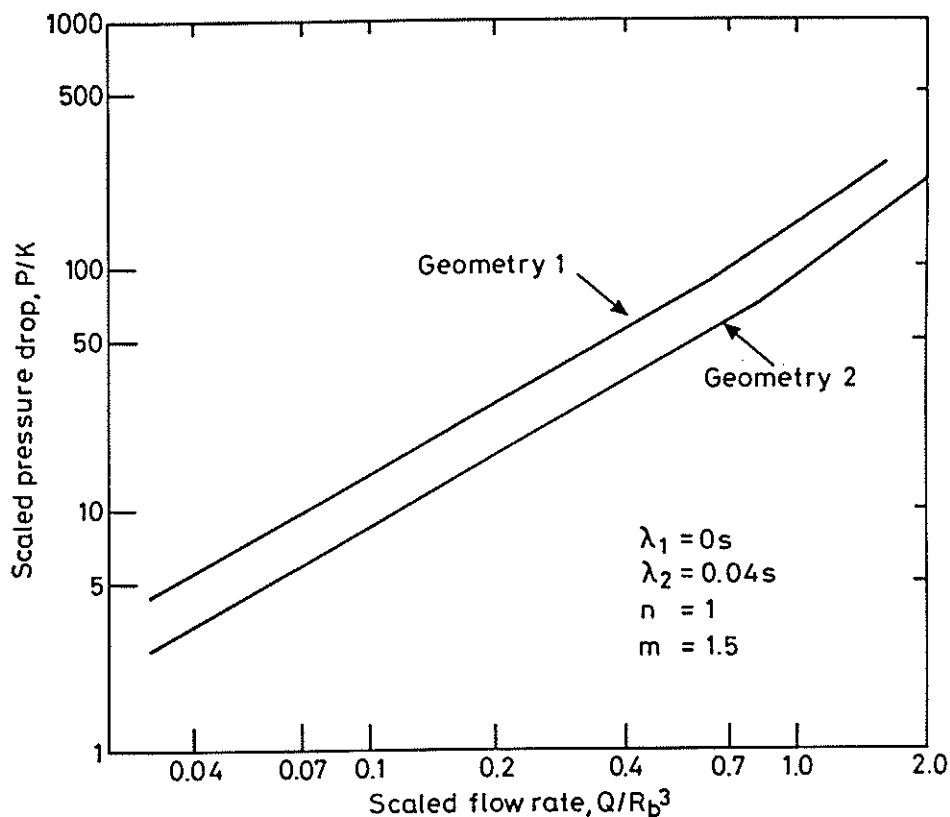


Fig 11. Comparison of calculated entrance pressure drop values for Geometries 1 and 2.

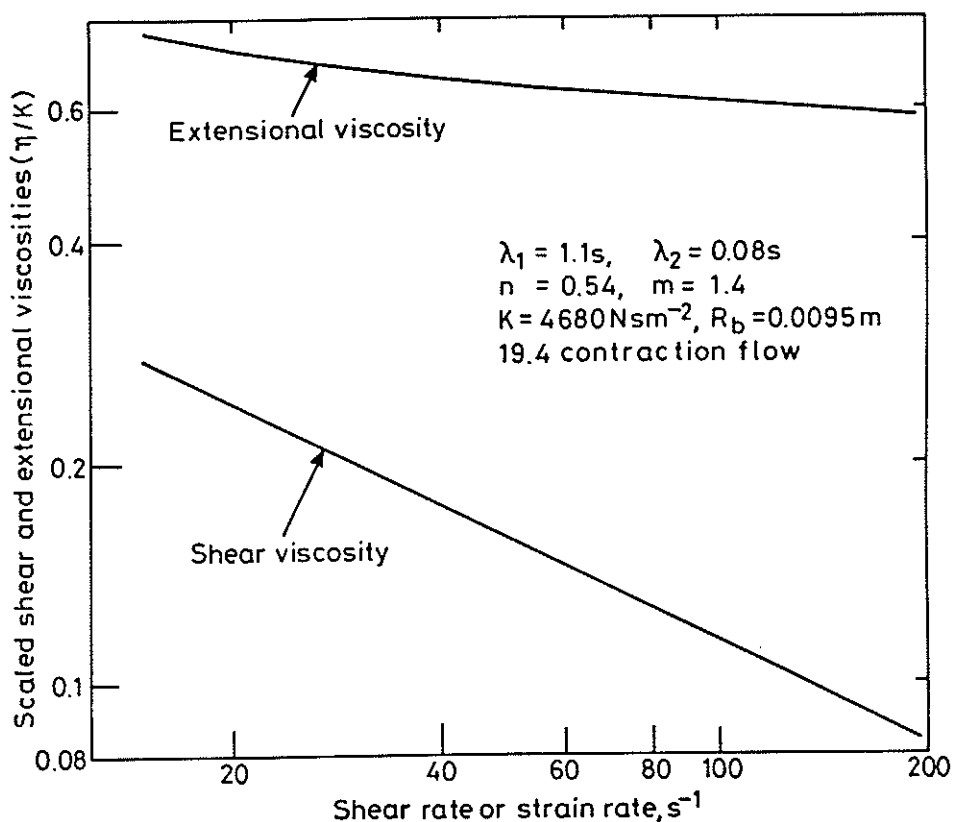


Fig 12. Calculated spectral model extensional viscosity values determined from capillary rheometry entrance pressure drop versus flow rate data using the numerical array, with measured DMC slurry shear viscosity data presented for comparison.

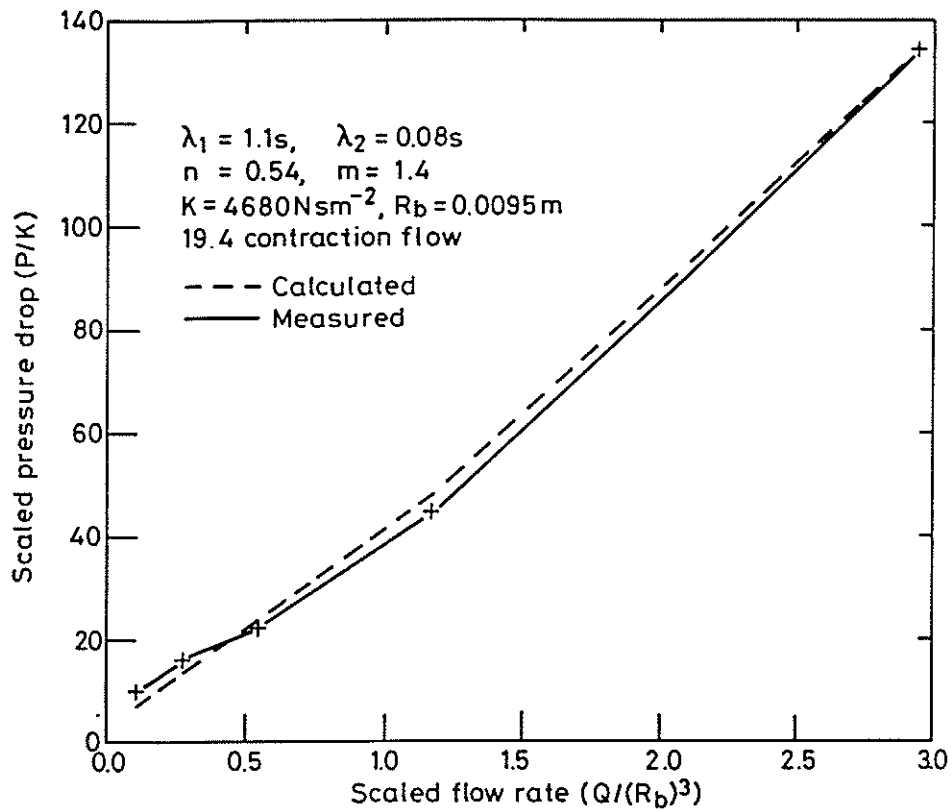


Fig 13. Comparison of measured and calculated entrance pressure drop curves for a DMC slurry, demonstrating the reliability of the method used.

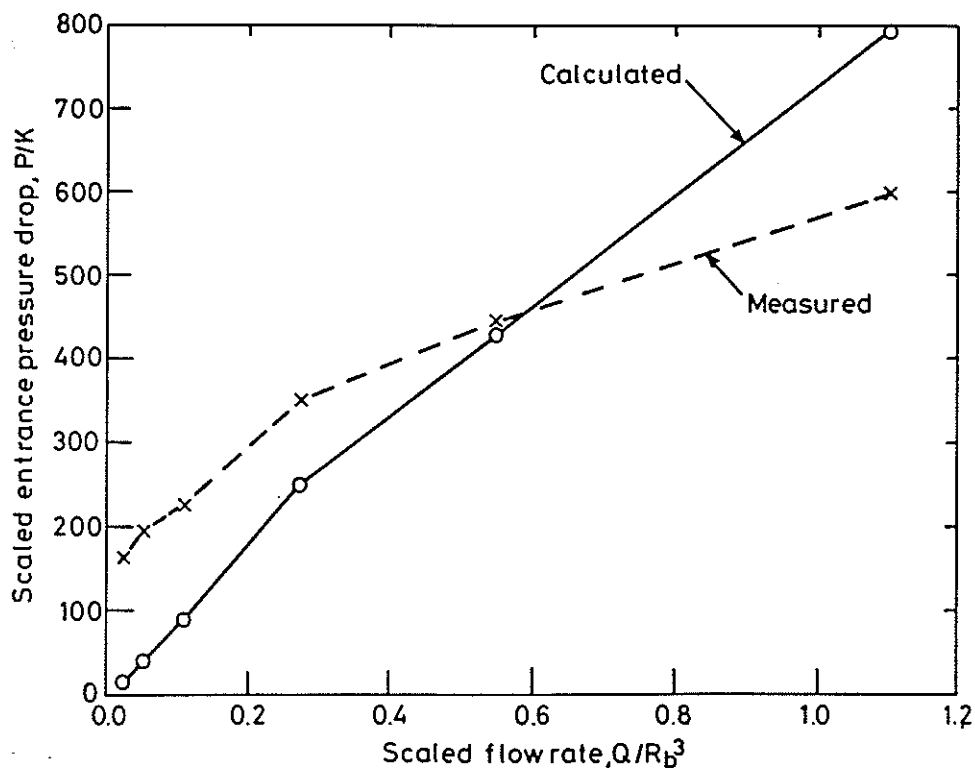


Fig 14. Comparison of the measured and calculated entrance pressure drop curves for a glass-fibre filled unsaturated polyester DMC using a 19:4 diameter contraction ratio ($R_b = 0.0095 \text{ m}$) and $\lambda_1 = 0.1 \text{ s}$, $n = 0.4$, $\lambda_2 = 202 \text{ s}$, $m = 1.17$ and $K = 3320 \text{ Nsm}^{-2}$.

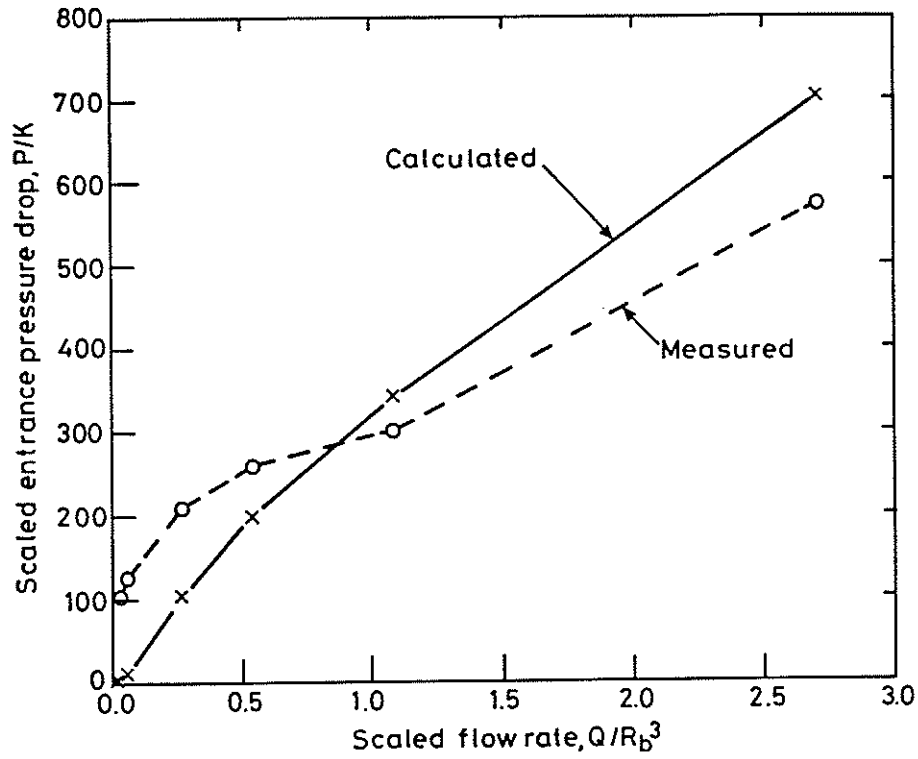


Fig 15.

Comparison of the measured and calculated entrance pressure drop curves for a glass-fibre filled unsaturated polyester DMC using a 19:6 diameter contraction ratio ($R_b = 0.0095$ m) and $\lambda_1 = 0.1$ s, $n = 0.4$, $\lambda_2 = 202$ s, $m = 1.17$ and $K = 3320$ Nsm^{-2} .