Towards the Characterization of Viscoelastic Materials Using Model-Based Reasoning Techniques

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Abstract

A rheological equation of state is required to simulate the behaviour of a viscoelastic material in many complicated industrial flow processes. The generation of such a model is not a straightforward operation. In this paper we show how this process may be automated using model-based reasoning techniques. Data from three simple experiments are required in our analysis in order to completely characterize a material and formulate its equation of state.

This paper builds on the work in (Capelo, Ironi & Tentoni 1992; 1993) who have presented a system which selects a mathematical model (the constitutive equation) for a given viscoelastic material from its response to an externally applied force in a so-called creep-test. This process is based on a qualitative abstraction of the plots generated from this static experiment.

We extend this work to fully characterize the material in terms of its rheological constants and functions. The viscosity is then determined from a steady simple shear experiment in which the data is analyzed using a graph recognition technique. The relaxation and retardation times are determined using data from a dynamic experiment. An optimization technique is used to determine the number of relaxation and retardation times and the values of these parameters. This system delivers to the user a constitutive relationship with material constants and functions which may be used to simulate the flow of the viscoelastic material under consideration.

Introduction

An understanding of the behaviour and properties of viscoelastic materials, whose ranks include materials such as paints, shampoo, polymers and lubricants, is important in many industrial processes. Many important and industrially relevant questions remain unanswered. For example, in journal bearing lubrication, can normal stresses induced by viscoelasticity compensate for the adverse effects of shear-thinning, namely higher wear resulting from lower friction. The formulation of a realistic viscoelastic model enables the behaviour of a given lubricant to be predicted by means of numerical simulation. The mathematical description of a viscoelastic fluid is much more complex than its Newtonian counterparts. In addition to the conservation equations of mass and momentum an additional equation, the constitutive equation or rheological equation of state, is required which relates the stress to the deformation. For a viscoelastic liquid this relationship is nonlinear and it has no standard form which is universally valid for each fluid in every flow situation. This situation is one of the reasons why the subject of viscoelasticity is so challenging.

A constitutive equation is required in order to perform numerical simulations of viscoelastic flows. Although some information can be gleaned from experiments a complete description can only be obtained by solving the full set of governing equations. This enables local characteristics such as information about the stresses and the energy dissipation, for example, to be calculated. A constitutive model which is simple enough to allow efficient numerical solution yet possessing predictive capabilities needs to formulated.

In practice the constitutive equation is formulated by appealing to rheometrical experiments. The behaviour of the material is investigated in simple flows such as steady simple shear flow and small amplitude oscillatory shear flow. Then a model, which is able to simulate the behaviour of the fluid in these simple flows, is chosen from among those that are available. The work in this paper automates much of this process generating a complete constitutive equation directly from experimental observations.

In this paper we present an intelligent hybrid system which automates both the selection of the form of the constitutive equation and the determination of the material constants and functions which appear in this equation. In (Capelo, Ironi & Tentoni 1992; 1993) a system is presented which selects a mathematical model (the constitutive equation) for a given viscoelastic material from its response to an externally applied force in a so-called creep-test. This process is based on a qualitative abstraction of the graphical output generated from this static experiment. Their analysis shows that there are four physically feasible classes of models. Each class contains a family of models each member being identified by the number of relaxation times of the material. At this stage the work of calculating the material functions and parameters of the constitutive equation (1) and also automating the process of determining the number of relaxation or retardation times (constant terms on the left and right hand side, respectively) has not yet been done.

Therefore, in this paper we extend their work by building an intelligent hybrid system which can fully characterize the material in terms of its rheological constants and functions. A selection of rheometrical experiments are necessary to provide data for this procedure. Once the equivalence class for the material has been identified the material constants and functions may be determined. The viscosity is determined from a steady simple shear (viscometric) experiment in which the data is analyzed using a graph recognition technique developed by (Mustapha et al 1997). The relaxation and retardation times are determined using data from a dynamic experiment. An optimization technique is used to determine the number of relaxation and retardation times and the values of these parameters. This system delivers to the user a constitutive relationship complete with values of the material constants and functions which may be used to simulate the flow of the viscoelastic material under consideration.

Formulation of a Rheological Model from a General Constitutive Equation

Good progress towards the formulation of a rheological model from given experimental data has been made by (Capelo, Ironi & Tentoni 1991; 1992; 1993; 1995). There are four basic processes which enable them to formulate a general rheological equation of state. These are summarized in Figure 1.

A static creep test experiment is performed on a given viscoelastic material and the strain response is analyzed. This analysis is performed using graph recognition techniques. In (Capelo, Ironi & Tentoni 1993) this information is used to assign to this material one of four formal admissible constitutive equations:

$$\sigma + \sum_{i=1}^{m} \alpha_i D^i \sigma = \sum_{i=\delta}^{n} \beta_i D^i \gamma, \qquad (1)$$

where $n = m, m+1$, $\delta = 0, 1$, α_i, $i = 1, \ldots, m$, β_i, $i = \delta, \ldots, n$ are material functions and D is the time derivative operator. The function β_1 is the viscosity and we will also denote this by η. All the other material functions in (1) are assumed to be constant.

Process I shows a segmentation of the strain response based on several successive steps of stress imposed on the material. Therefore the segmented components are quite clear. Excellent descriptions for each of the components can be found in (Whorlow 1980). Using first and second order derivatives, the qualitative curve attributes can be obtained (Capelo, Ironi & Tentoni 1995). A full qualitative description such as instantaneous, delayed elasticity and non-recoverable deformation can be obtained by applying heuristic rules as shown in Process II. Consequently, in Process III, if the existence of certain properties such as instantaneous, delayed elasticity and full recovery (non-viscosity) are realised, then the value of the logical triplet for this example is (T,T,F). The other three possibilities are (F,T,T), (F,T,F) or (T,T,T). Process IV shows that a direct mapping to an equivalence class is a straightforward task once the logical triplet is obtained.

The two major drawbacks in the procedure described above are : *i) the number of relaxation or retardation times are manually input from user and ii) the material parameters or functions such as the viscosity, relaxation and retardation times are unavailable.* To overcome these difficulties, we propose two further experiments to fully characterize the material. Thus, three experiments in all will suffice to determine a constitutive equation for a given viscoelastic material. The interaction between the output from these three experiments forms the basis of the intelligent hybrid system shown in figure 2. The additional experiments are:

1. *Dynamic experiment :* Perform a small-amplitude oscillatory shear experiment. The outputs from this experiment are the storage and loss moduli denoted by G' and G'' respectively. The discrete relaxation spectra can be determined from this data.
2. *Viscometric experiment :* If required, run a steady simple shear experiment to determine the shear viscosity.

The details which show the output of the viscometric data analyser (calculating viscosity) can be found in (Mustapha 1995; Mustapha et al 1997). Under the assumptions that the fluid is a linear viscoelastic liquid, the flow conditions are isothermal and extensional viscosity is considered small, the system shown in figure 2 is a complete one. Section 3 will explain in detail the calculation of a viscosity function from data obtained from a simple steady shear experiment. Section 4 shows how the discrete relaxation spectrum of a material may be calculated.

Determination of the Viscosity Function

The most important material property which needs to be determined is the viscosity. The flow of the material in many situations can be predicted from a knowledge of this function. The viscosity can depend on variables such as shear rate, temperature and pressure. However in many industrial and everyday situations it is the variation of viscosity with shear rate which is the most important. Therefore in this paper we restrict ourselves to this situation. Several models are available for the purpose of fitting the viscosity-shear rate data and each of them has its own capabilities in characterising different shapes of the flow curve. In the system developed by (Mustapha 1995; Mustapha et al 1997), six models are used. The process of determining several models on a piecewise rheogram is found in two stages :-

1. Explanation process (qualitative technique) : It is important to explain to the user the meaning of the curve in order to help choose the piecewise curve (up or down curve) and the range of the curve chosen (initial or endmost or whole part of the curve). The meaning of the mathematical models are also necessary since the user needs to know why and when to use them. In additional to these there are warning messages for graph abnormalities and expert advice to repeat the experiment using the proper geometry. Since only a qualitative description is required to fulfil these tasks, qualitative reasoning techniques are the most appropriate.

2. Curve-fitting process (quantitative technique) : The objective of this curve-fitting process is to find the best model/s and also to minimise the number of models used in characterising the selected curve. Frequently there will be no single model which is capable of characterizing the material over the whole shear rate range. The criteria for selecting the models are as follows :-

   (a) The model must cover the largest shear rate range. This is important in minimizing the number of models used.

   (b) The model must produce the highest correlation among all other models or at least above 0.9975. The correlation is obtained by Pearson's method (Weiss & Hassett 1991).

The six models currently used in the system are listed in Table 1 where τ is the shear stress and $\dot{\gamma}$ is the shear rate. Figure 3 illustrates how several models can be assigned to a flow curve in a piecewise sense.

Determination of the Discrete Relaxation Spectrum

Suppose that following the creep test we have identified which of the four admissable constitutive equations describes a given viscoelastic material. In this general equation the number of relaxation times (the value m in (1)) needs to be determined in order to reduce the number of plausible models further. Ideally we would like to describe the rheological behaviour of the material with as few parameters as possible for two important reasons. First of all to reduce the test of numerical simulations and secondly to avoid the ill-posed problems associated with the determination of the discrete relaxation spectra of a viscoelastic material. This is well-known to be an ill-posed problem in which the degree of ill-posedness increases as the number of relaxation times increases (Honerkamp 1989).

The relaxation modulus $\phi(t)$, defined by

$$\phi(t) = \sum_{j=1}^{m} g_j \exp(-t/\lambda_j), \quad t \geq 0,$$

may be measured using dynamic mechanical methods in which the material is strained sinusoidally at a frequency ω. The measured stress response decomposes into an in-phase and an out-of-phase component: the storage modulus G' and the loss modulus G''. In oscillatory shear we define a complex shear modulus G^* through the equation

$$\sigma(t) \; = \; G^*(\omega)\gamma(t). \tag{2}$$

The complex shear modulus, $G^*(\omega)$, has the representation

$$G^*(\omega) = \int_0^\infty \phi(s)\exp(-i\omega s)ds = \sum_{j=1}^{m} \frac{i\omega g_j \lambda_j}{(1+i\omega\lambda_j)}.$$

We write

$$G^*(\omega) = G'(\omega) + iG''(\omega),$$

where

$$G'(\omega) \; = \; \sum_{j=1}^{m} \frac{g_j \lambda_j^2 \omega^2}{(1+\omega^2\lambda_j^2)},$$

$$G''(\omega) \; = \; \sum_{j=1}^{m} \frac{g_j \lambda_j \omega}{(1+\omega^2\lambda_j^2)},$$

Experimental data provides us with information at $\omega = \omega_i$, $i = 1, \ldots, M$. Let us denote this by G'_i and G''_i, $i = 1, \ldots, M$, respectively. This results in the following system of equations for g_j, λ_j, $j = 1, \ldots, m$:

$$G'_i \; = \; \sum_{j=1}^{m} \frac{g_j \lambda_j^2 \omega_i^2}{(1+\omega_i^2\lambda_j^2)},$$

$$G''_i \; = \; \sum_{j=1}^{m} \frac{g_j \lambda_j \omega_i}{(1+\omega_i^2\lambda_j^2)},$$

for $i = 1, \ldots, M$. This is a system of $2M$ equations for $2m$ unknowns if m is known. However, the value of m is not known priori and must be determined along with the other parameters to avoid unreliable numerical simulations at a later stage. The nonlinear nature of this system of equations makes it extremely difficult to solve. One has to resort to a nonlinear least squares method in order to fit the data.

We consider the minimization of the function

$$\chi^2 = \sum_{i=1}^{M} \left\{ G_i' - \sum_{j=1}^{m} K_{i,j}' g_j \right\}^2 \frac{1}{(\sigma_i')^2} + \sum_{i=1}^{M} \left\{ G_i'' - \sum_{j=1}^{m} K_{i,j}'' g_j \right\}^2 \frac{1}{(\sigma_i'')^2},$$

with respect to m, λ_j and g_j, $j = 1, \ldots, m$, where

$$K_{i,j}' = \sum_{j=1}^{m} \frac{\lambda_j^2 \omega_i^2}{(1 + \omega_i^2 \lambda_j^2)},$$

$$K_{i,j}'' = \sum_{j=1}^{m} \frac{\lambda_j \omega_i}{(1 + \omega_i^2 \lambda_j^2)}.$$

The measured values $G_i^{'}$ and $G_i^{''}$ are assumed to be affected by standard errors of size $\sigma_i^{'}$ and $\sigma_i^{''}$, respectively. The algebraic equations resulting from the least squares process are solved using the Marquardt-Levenberg algorithm. This turns out to be a highly efficient numerical algorithm for this problem. The Marquardt-Levenberg algorithm is a standard optimization procedure although to our knowledge it has not been used previously in this context.

The number of relaxation modes is increased gradually until there is no improvement in the minimization of χ^2. There are a number of criteria which may be applied in order to terminate the procedure before the problem becomes ill-posed. One termination criterion is to stop when negative values of the parameters appear since these are physically infeasible. However, realising that this criterion is insufficient, one of the future tasks is to work out a more robust termination criterion.

Table 2 shows a sample 6-mode relaxation spectrum generated for a polymer melt and a decrease of standard errors as the number of modes (i) increases. The corresponding result in Table 2 can be shown graphically in Figure 4. Given a discrete relaxation spectrum the corresponding discrete retardation spectrum may be determined using the Laplace transform (Weiss & Hassett 1991).

Conclusions

We have built on the work in (Capelo, Ironi & Tentoni 1991; 1992; 1993; 1995) and used data from additional experiments in order to fully characterize a given viscoelastic material. The viscosity is determined from a steady simple shear experiment in which the data is interpreted using qualitative and quantitative techniques. The discrete relaxation spectrum is determined from a small-amplitude oscillatory shear experiment. The extraction of the relaxation spectrum from this data is an ill-posed problem. A nonlinear least squares method is proposed for circumventing the problems associated with ill-posedness. This method also determines the appropriate number of relaxation times for the material.

One limitation of this work is that we have assumed that viscosity is a function of shear rate only. In practice it can also depend on pressure and in a non-isothermal setting, temperature.

The methods described in this paper have provided good results from rheological experiments with a variety of substances.

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Viscosity model	*Equations*	*Meaning of model parameters*
Newtonian model	$\eta_0 = \frac{\tau}{\dot{\gamma}}$	η_0 - viscosity at zero-shear
Bingham model	$\eta = \frac{\tau_y}{\dot{\gamma}} + \eta_p$	η_p - plastic viscosity τ_y - yield stress
Sisko model	$\eta = \eta_\infty + k\dot{\gamma}^{n-1}$	η - viscosity η_∞ - infinite-rate viscosity k - viscosity coefficient or consistency n - rate index
Casson model	$\eta^{1/2} = \dot{\gamma}^{1/2}\tau_y^{1/2} + k\dot{\gamma}$	k - consistency or viscosity coefficient τ_y - yield stress
Power Law	$\eta = k\dot{\gamma}^{n-1}$	k - consistency or viscosity coefficient n - Power Law index
Herschel-Bulkley	$\eta = \frac{\tau_s}{\dot{\gamma}} + k\dot{\gamma}^{n-1}$	τ_s - yield stress k - consistency or viscosity coefficient n - Power Law index

Table 1: *Viscosity models*

i	λ_i	g_i	σ
1	28.4075	4490	8.7618e-01
2	3.0371	7679	4.5586e-01
3	0.5184	18400	1.9621e-01
4	0.0911	38828	7.9216e-02
5	0.0164	74196	3.3643e-02
6	0.0023	167850	1.8901e-02

Table 2: *Relaxation spectrum for a polymer melt*

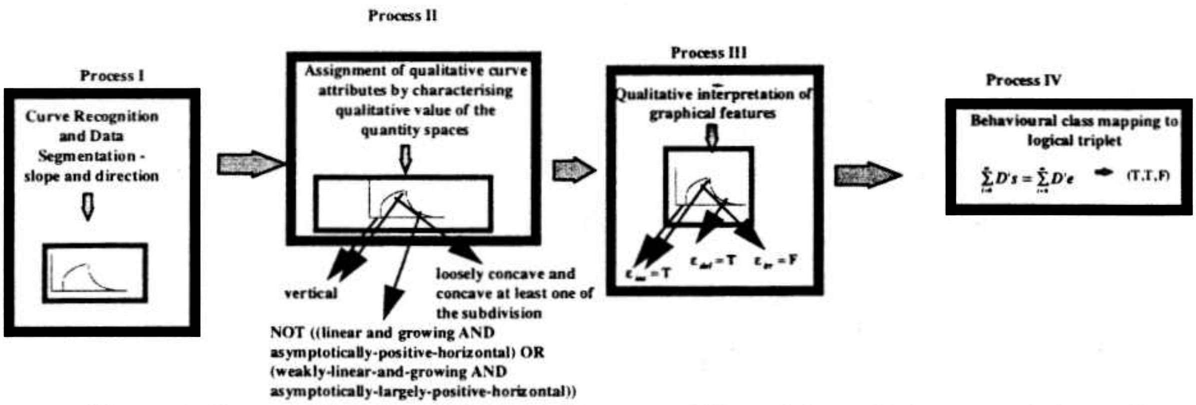

Figure 1: *Four major processes in analysing creep data and determining an equivalence class*

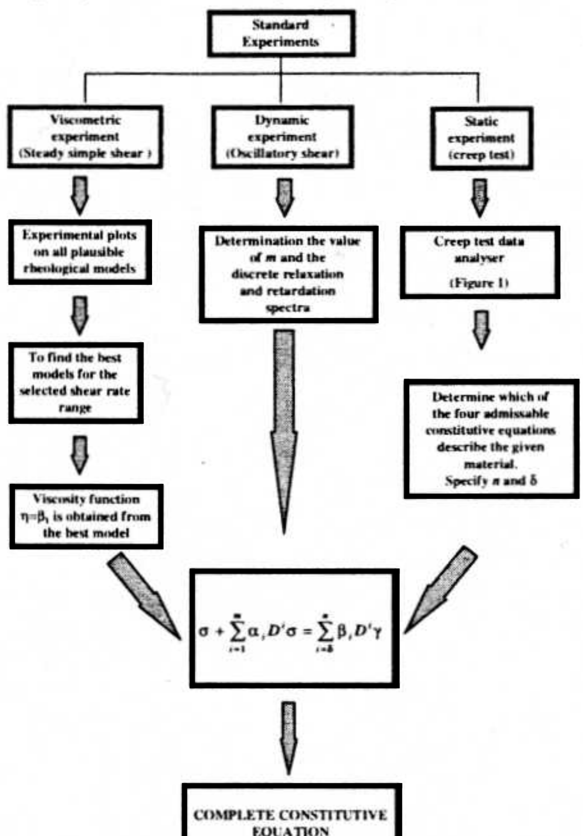

Figure 2: *An intelligent hybrid system for determining a complete rheological model for a viscoelastic material*

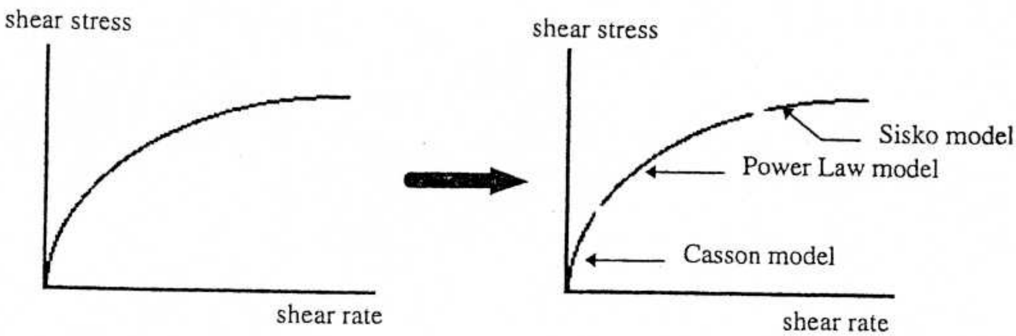

Figure 3: *A flow curve which has models assigned to it*

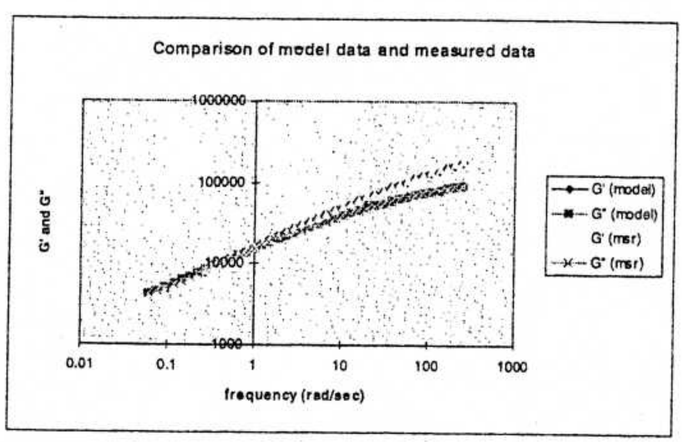

Figure 4: *A comparison of model data and measured data for polymer melt*