PURE MATERIAL INSTABILITY AND THE CONCEPT OF YIELD STRESS

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ABSTRACT

The paper is concerned with the rheological study of gels, complex materials which are characterised by an internal network structure developed in viscous liquids. This category of materials exhibits in viscometric motion a yield state at a critical value of the applied shear stress. The yield shear stress defines the plateau behaviour in the steady flow curve. The creeping experiments and the dynamics of the Oldroyd's 3 constants model put in evidence the connection between the loss of stability of the network structure and the coexisting strain rates at a constant shear stress. The correlation between theory, numerical simulations and experiments are established. All the results are qualitatively consistent with the statement that the concept of yield stress is 'natural defined' in the context of the pure material (structural) instability (i.e. instability at zero Reynolds number) of constitutive relations with non-monotone flow curve.

KURZFASSUNG

In dieser Arbeit wird das rheologische Verhalten der Gelen untersucht, d.h. viskoser Materialien, die eine netzartige Struktur aufweisen, welsche sich im Inneren einer viskosen Flüssigkeit entwickelt. Diese Klasse von Materialien wird im den viskometrischen Strömungen durch eine Fließgrenze charakterisiert, welche einem kritischen Wert der Schubspannung entspricht. Die Schubspannung bestimmt das Plateauverhalten der stationären Fließkurve. Die unter einer konstanten Schubspannung durchgeführten Versuche und die Dynamik des Oldroyd-Modells mit 3 Materialkonstanten zeigen den Zusammenhang zwischen dem Verlust der Stabilität der Netzstruktur und den auftretenden Deformationsgeschwindigkeiten bei konstanter Schubspannung. Die Korrelation zwischen Theorie, numerische Simulation und Experiment wird festgelegt. Alle Resultate stehen in qualitativer Übereinstimmung mit der Annahme, daß der Begriff 'Fließgrenze' im Zusammenhang mit der puren Materialinstabilität (d.h., bei verschwindender Reynolds-Zahl) der Materialgleichungen mit nichtmonotoner Fließkurve 'natürlich' erklärt werden kann.

RÉSUMÉ

Dans ce travail on étudie le comportement rhéologique des gels: des matériaux viscoélastiques caractérisés par une structure du type de réseau (network), développée dans un liquide visqueux. Cette catégorie de matériaux est caractérisée, dans les mouvements viscométriques, par un seuil d'écoulement. Le seuil d'écoulement definit le comportement de plateau dans l'écoulement stationnaire. Les expériences développées sous l'action d'une contrainte tangentielle constante, ainsi que la dynamique du modèle d'Oldroyd à trois constantes de matériel, mettent en évidence la liaison entre la perte de la stabilité du réseau et la coexistence de quelques vitesses de déformation spécifiques différentes à une contrainte tangentielle constante. On a établit la corrélation entre la théorie, les simulations numériques et les expériences. Tous les résultats sont consistants du point de vue qualitatif, avec l'affirmation que le concept de seuil d'écoulement est définit d'une maniere naturelle dans le contexte de l'instabilité purement matérielle (l'instabilité au nombre de Reynolds égal à zéro) des relations constitives à courbe d'écoulement non monotone.

KEY WORDS:

material instability, yield stress, network structure, creeping motion, dynamic behaviour, gels, non-monotone flow curve.

1 INTRODUCTION

Complex viscoelastic materials generic called "gels", concentrated dispersed systems in viscous fluids, develop a stable entanglement or cross-linked network structure at rest, structure which is destroyed during the shearing beyond a critical value of the applied shear stress. This value is associated with the yield stress τ_o, respectively with the plateau behaviour of the steady flow curve and the sharp decreasing of the viscosity function in viscometric motions, see Fig. 1. In this category of materials we can include: colloidal and flocculated suspensions (Coussot *et al.* 1993; Mas and Magnin, 1993), liquid crystals (Olmsted and Goldbart, 1992), suspensions in polymer solutions (Kawaguchi, 1994), biopolymer gels (Lapasin and Pricl, 1998), electrorheological fluids (Marshall *et al.* 1989, Pan and McKinley, 1997)

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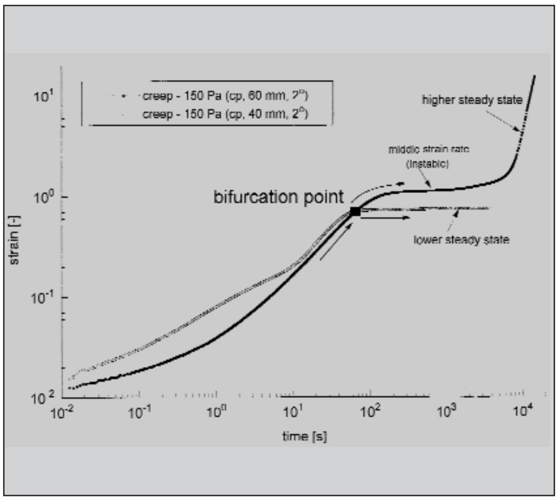

Figure 6 (left): The experimental dependence $\gamma(t)$; lubricating grease at 20° C, TA controlled stress rheometer AR 1000 N(University of Wales at Aberystwyth).

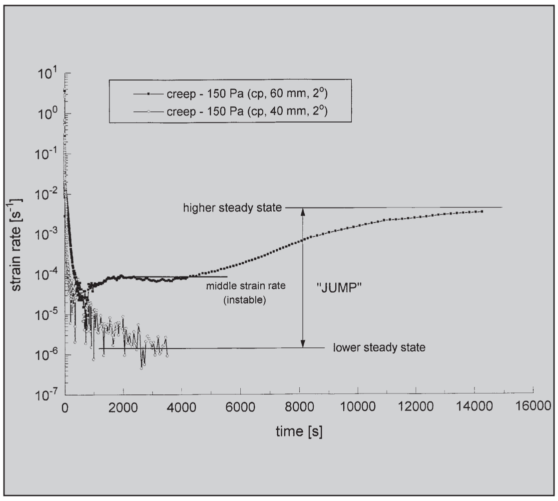

Figure 7 (right): The experimental dependence, $\dot{\gamma}(t)$; lubricating grease at 20° C.

in the absence of external perturbations, the solution $De(t)$ is attracted to one or the other stable steady solutions $De_1 = \lambda_1\dot{\gamma}_1$, respectively $De_2 = \lambda_1\dot{\gamma}_2$, as a function of the initial values $De(0)$ and $D\dot{e}(0)$, for details of the dynamics of differential equations see Rüdiger (1994). The complete dynamics of the equation (5) is studied in [1].

The solutions of Eq. 5: (i) strain dependence $\gamma(t)$, (ii) strain rate dependence $De(t)$ and (iii) the parametric time dependence $De\,(D\dot{e})$, are shown for the Jaumann derivative in Figs. 5 at $\overline{\tau}(0) = 0.4$, $De(0) = 50$ and three different values of $D\dot{e}(0)$.

The first steady solution De_1 is always reached in a monotone way (Fig. 5c), whereas high oscillations are observed for De_2 (Figs. 5a - 5b). For a certain range of the ratio $De(0)/D\dot{e}(0)$ is possible to reach in one single numerical experiment both solutions (Fig. 5b).

The creep experiments were performed with commercial lubricating greases at constant temperature (this material is considered to be representative for the gels family). Two cone and plate configurations have been used: the cones diameters of 40 mm and 60 mm, with the angle between the cones and the plate of 2 degrees. The constant torque is applied on the cone and the deformation of the sample is measured on the same surface. The strain rate has been directly computed as the time derivative of the strain. Neither fracture, ejection or real slipping at the contact surface between the sample and the cone have been observed during the experiments.

The experimental curves $\gamma(t)$, $\dot{\gamma}(t)$ and the corresponding parametric plot $\dot{\gamma}(\ddot{\gamma})$ at the constant shear stress $\tau = 150$ Pa are represented in Figs. 6, 7, 8. The dependence $\gamma(t)$ put in evidence the mechanism of the pure material instability. Both experiments are identically up to a bifurcation point. Then, one experiment is attracted by the lower steady state, whereas the other is attracted by the higher steady state. The middle strain rate is reach but the solution does not remain there due its non-stable character. The dynamics of the strain is direct related with the experimental relevance of the jump in strain rate from the lower to the higher steady state, through the instable strain rate, see Fig. 7.

The similarities with the numerical simulations shown in Figs. 5 are evident. Actually, there is no difference from qualitative point of view between the experiments and the dynamics of the constitutive relation (5). The lack of oscillations in the experiments could be explained by the influence of the term "$ReWi\,\partial\overline{v}/\partial\overline{t}$" from (1), which is acting as a damper for the material oscillations due the contrary effect on the flow field of the inertia in front of the material and elastic instabilities [8, 11].

4 *FINAL REMARKS*

In the limit of zero Reynolds number, we interpret the yield stress as the constant value of the shear stress for which a bifurcation point is present in the dynamic behaviour of the constitutive relation. This corresponds with the loss of stability of the network structure of the gels. Beside the parameters κ, a_1, a_2 the location of the bifurcation point is a function of the initial conditions of the Deborah number and its time derivative.

These initial conditions for the constitutive relation (5) are naturally imposed from the interaction between the constitutive relation and the equation of motion, through the boundary conditions. We are looking to the change in the flow geometry as an initial perturbation acting on the boundary of the flow domain. In our interpretation the values $De(0)$ and $D\dot{e}(0)$ are material parameters which define the instantaneous answer of the network structure in a particularly space

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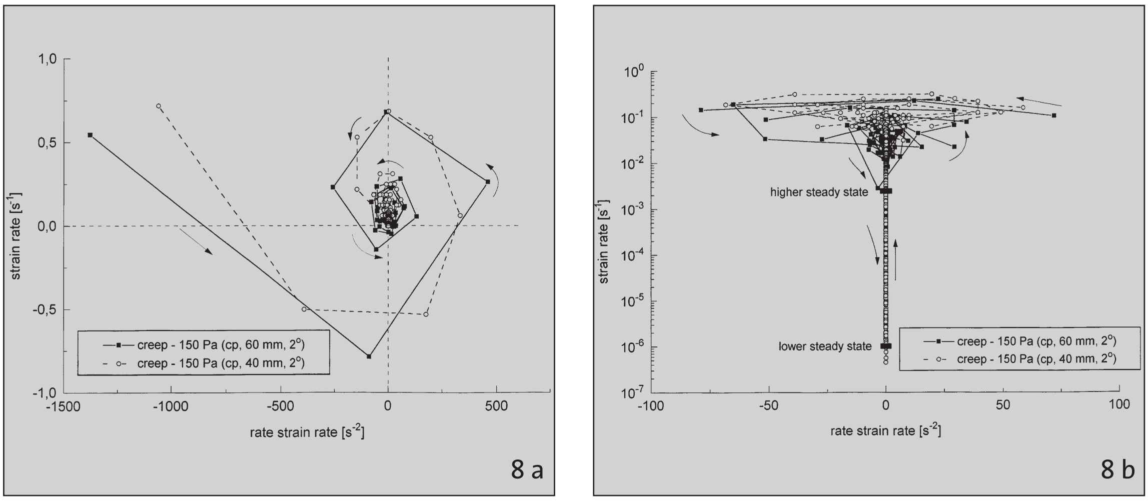

Figure 8:
The experimental dependence, $\dot{\gamma}(\ddot{\gamma})$;
with time as parameter;
a) normal scale,
b) logarithmic scale (detail);
lubricating grease at 20° C.

domain. The influence of the geometry in our experiments was expected, since it is well known the influence of the gap on the rheometry of these category of materials, see for details Barnes (1995). Actually, the answer of any "viscoelastic solid structure" is sensitive on the space scale of the motion, not only on the type of the load or the deformation process. Contrary, for structural stable materials (as pure viscous liquids, for example) the influence of the gap would not have been observed, at least in viscometric motions.

The developing of novel technological processes and complex materials belong to the family of gels are direct dependent on the understanding of the onset of their fluid behaviour. Further experimental investigation of the dynamics of creep motion are needed to find out a complete validation of the hypothesis and interpretations proposed in the paper. This is possible using a visualisation technique of the deformation field in the gap of the controlled stress rheometer and extending the experimental studies to the extensional motions. New experimental investigations has to be direct related with the theoretical modelling in order to produce fundamental knowledge on the rheological behaviour of these complex materials, as well as scientific explanations and support for concepts which are considered at this moment semi-empirical ones.

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BIOGRAPHY

Prof. Corneliu Balan was born in 1956 in Bucharest, Romania. He received a M.Sc. in Fluid Mechanics at "Politehnica" University and the Ph.D. degree in Continuum Mechanics at TH Darmstadt (Prof. K. Hutter). Post-doctoral studies in Continuum Mechanics and Rheology at University of Wales (1993) and University of Minnesota (1994/95).

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