

Normal Stress Measurements for Viscoelastic Liquids Using Real Time Holographic Interferometry of the Weissenberg Effect

M.F. HIBBERD

Post Graduate Student, Department of Physics, The Australian National University, Canberra

and

H.G. HORNING

Reader, Department of Physics, The Australian National University, Canberra

SUMMARY Earlier theoretical research has shown that the shape of the free surface near a rotating rod depends on the geometry of the apparatus and the elastic properties of the liquid. Some experimental work is described in which holographic interferometry is used to measure small variations in the free surface shape for spherically and cylindrically symmetric geometries. These results were analysed in terms of the above-mentioned theory to provide values of the two normal stress difference coefficients in the second order region of some simple viscoelastic liquids.

1 INTRODUCTION

There is a vast amount of literature dealing with measurements of the many rheological properties of viscoelastic materials. By restricting attention to steady isothermal flows of incompressible fluids, we consider only two of the simplest properties which are difficult to measure. For the flow of a Newtonian liquid, the only material properties required are the viscosity and density. For simple viscoelastic liquids we need to include a history of the shearing, albeit a fading memory of it. To simplify this history we consider only viscometric flows, in which each particle experiences a constant shear rate. For such flows it can be shown that the material properties required are the shear stress and the first and second normal stress differences N_1 and N_2 respectively. These are termed the viscometric functions and depend on the shear rate. N_1 is the difference in stress between the direction of flow and the direction normal to the shear planes, whereas N_2 is the difference in stress between the two directions normal to the flow.

The normal stress ratio N_2/N_1 is generally agreed to lie in the range -0.1 to -0.4 for moderately concentrated polymer solutions (Bird et al., 1977) but some experiments show a larger range. Correlations with temperature, concentration and molecular structure are unclear. Most difficulty is experienced measuring N_2 , but N_1 can also be difficult to measure for weakly elastic liquids. Although in many practical applications the smallness of N_2 means that it can be neglected, there are some flows where it is important and some recent molecular theories provide values for N_2/N_1 which need to be checked.

As an alternative to directly measuring pressures in the flowing fluid a number of workers have explored the use of the free surface shape as a measure of the state of stress in the fluid. Apart from Tanner's trough flow experiments (Kuo and Tanner, 1974), all free surface measurements seem to have been of the Weissenberg effect (rod climbing) in cylindrical symmetry. Most of these are mentioned by Beavers and Joseph (1979).

Böhme (1974) has shown that for sufficiently slow flow it is possible to obtain explicit expressions for the shape of the free surface of a liquid filling the gap between rotating solid surfaces of revolution. The present work uses the sensitivity

of holographic interferometry to measure the small changes resulting from the slow flow and thus obtain surface profiles at various speeds of rotation of the inner surface. The theory is fitted to these results to provide separate values for N_1 and N_2 at low shear rates.

2 THEORY

We consider steady isothermal flow of an incompressible fluid with velocity vector $\mathbf{v} = (\dot{\gamma}x_2, 0, 0)$ in a Cartesian coordinate system with position vector (x_1, x_2, x_3) and shear rate $\dot{\gamma}$. For a simple second order fluid at small shear rates we can then write the viscometric functions in terms of the non-zero components of stress σ_{11} , σ_{22} , σ_{33} and σ_{12} as

$$\sigma(\dot{\gamma}) = \sigma_{12} = \eta_0 \dot{\gamma} + O(\dot{\gamma}^3)$$

$$N_1(\dot{\gamma}) = \sigma_{11} - \sigma_{22} = \Psi_{1,0} \cdot \dot{\gamma}^2 + O(\dot{\gamma}^4) \quad \dots(1)$$

$$N_2(\dot{\gamma}) = \sigma_{22} - \sigma_{33} = \Psi_{2,0} \cdot \dot{\gamma}^2 + O(\dot{\gamma}^4)$$

where $\Psi_{1,0}$ and $\Psi_{2,0}$ are called the zero shear rate first and second normal stress difference coefficients. Attention is restricted to "slow" flows defined by the conditions of small Reynolds and Weissenberg numbers,

$$Re = \rho V L / \eta_0 \ll 1 \quad \dots(2)$$

$$We = (|\Psi_{1,0}| + |\Psi_{2,0}|) \dot{\gamma} / \eta_0 \ll 1$$

where ρ is the fluid density and V and L are characteristic velocity and length scales. The Weissenberg number is the ratio of a characteristic time of the fluid to a characteristic time for the flow. Böhme (1975) examined such a flow between two rigid surfaces of revolution rotating with different small angular velocities about their common axis. He considered the flow to be made up of a primary creeping flow characterised by circular streamlines perpendicular to the axis of rotation. This induces a field of inertial forces and normal stresses which generally cause a secondary flow. The streamfunctions thus obtained were combined with particular boundary conditions to give explicit expressions for the shape of the free surface. Geometries considered in this paper are (i) coaxial circular cylinders with the inner one rotating and (ii) half-filled concentric spheres with the inner sphere rotating about an axis perpendicular to the fluid surface. The expressions for the surface shape Z are of the

form

$$Z(R) = g \cdot h(R) / \Omega_1^2 = A(R) \cdot \psi_{1,0} + B(R) \cdot \psi_{2,0} + C(R) \dots (3)$$

where $R = r/R_1$, r is the polar radius, Ω_1 and R_1 are the angular velocity and radius of the stirrer, and h is the change in surface height due to stirring. A, B and C are complicated functions of R with coefficients dependent on ρ , R_1 and the bowl radius R_2 ; they have different forms in spherical and cylindrical symmetry and in the cylindrical case $B(R) = 4 \cdot A(R)$. Thus separate values of $\psi_{1,0}$ and $\psi_{2,0}$ can be obtained from spherical surface shapes and checked with results for $(\psi_{1,0} + 4 \cdot \psi_{2,0})$ in cylindrical symmetry. The cylindrical theory assumes semi-infinite cylinders and so end effects must be investigated experimentally.

3 EXPERIMENTAL

3.1 Holographic Interferometry

In conventional interferometry it is necessary to have a reference object separate to the object under investigation. Holography allows one to store a 3-D image of the object in its undisturbed state and compare it at some later time with a slightly distorted version of the same object. A hologram (Collier et al., 1971) is essentially a recording of the interference between two light beams which are derived from a single coherent monochromatic light source. In our experiments the two beams are obtained using a variable ratio beam splitter at the output of a 100mW He-Ne laser. Each beam is expanded through a spatial filter to improve its spatial coherence. When recording a hologram, one beam illuminates the object and is scattered onto a photographic plate, the other (reference) beam illuminates the plate directly. If the plate is developed and reilluminated by the reference beam, the light scattered from the plate produces a 3-D virtual image of the original object.

In the actual experiments a hologram is taken of the bottom of the bowl through the unstirred viscoelastic liquid being investigated. After development the hologram is replaced in the same position in which it was exposed (within $\pm 0.5 \mu\text{m}$) using a simple six point-contact plate holder. When the hologram and the bottom of the bowl are initially reilluminated the virtual image of the object and the object itself coincide exactly. If the liquid is then stirred, there will be changes in the optical path length to the bottom of the bowl (due to the different refractive indices of the air and the liquid). The two images of the bottom of the bowl (one real, one virtual) will then be at slightly different positions and so interference fringes will be observed. For an illuminating direction near normal to the fluid surface a change in fringe number of m will be related to the actual change in surface height h by an equation of the form $h \approx \lambda m / (n-1)$ where λ is the wavelength of the illuminating light and n is the refractive index of the fluid. Changes in surface height in the range $0.3 \mu\text{m}$ to $60 \mu\text{m}$ can be resolved using this technique.

3.2 Apparatus and Procedure

The sensitivity of the fluid surface and holographic setup to building noise presented a major problem. For this reason all the experiments are performed on a 1.6m by 2.7m reinforced concrete table of webbed construction weighing $1\frac{1}{2}$ tons. It is supported on five inflated car inner tubes connected via capillary restrictions to larger air reservoirs to provide optimum damping and a resonant frequency of about

1 Hz.

The fluid stirrer consists of a base on which the spherical (or cylindrical) bowl is accurately located. The stirring rod, which has been ground to a fine finish, runs directly on lightly oiled nylon bushes pressed into two bearing plates which are supported above the bowl by three posts. The lower bearing plate also supports a front-silvered mirror which turns the illuminating beam through 90° . The runout of the stirring rod is less than $0.5 \mu\text{m}$, the axis of rotation is within 1 milliradian of the gravity vector and the axes of the rod and bowl are closer than 0.05mm . The radius of both bowls is 74.50mm , the inner sphere radius is 9.00mm and cylindrical rod radius is 9.29mm .

The drive system consists of a small sewing machine motor driven with a stabilised variable D.C. power supply. It drives a 300mm diameter pulley on the stirring rod via a 1.6mm diameter neoprene belt. The stirring speed is measured to $\pm 0.02 \text{ rad/sec}$ using an electronic circuit to count holes around the periphery of the main pulley. Typical stirring speeds range from 0.5 to 10 rad/sec.

Holograms are recorded with exposures of about 2 seconds on Agfa-Gevaert 8E75 plates. Photographs of the fringe patterns are taken over as large a range of stirring speeds as possible using a 200mm f4 lens and Kodak Plus X film developed for maximum contrast. The fringe contrast is quite high. Resolution is limited by speckle arising because of the use of diffuse reflections, and sometimes by noise in the stirrer disturbing the surface.

3.3 Data Analysis

The data is obtained as a series of fringe photos such as that shown in fig. 1. For various experi-

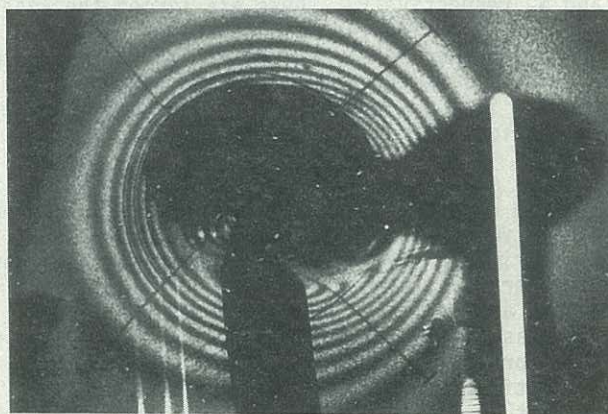


Figure 1 Typical holographic interferogram

mental reasons the illuminating beam is slightly divergent and the illuminating and viewing directions are not normal to the fluid surface. This complicates the interpretation of the fringe pattern, as does the actual method in which the fringes are formed which is more complicated than in conventional interferometry. A computer program is used to find the true surface profile using input data of fringe positions and various parameters describing the experimental setup. The program also computes values of $\psi_{1,0}$ and $\psi_{2,0}$ by fitting expressions of the form of equation 3 to the data using a least squares fitting technique.

4 RESULTS AND DISCUSSION

An initial check of the system was made with a light

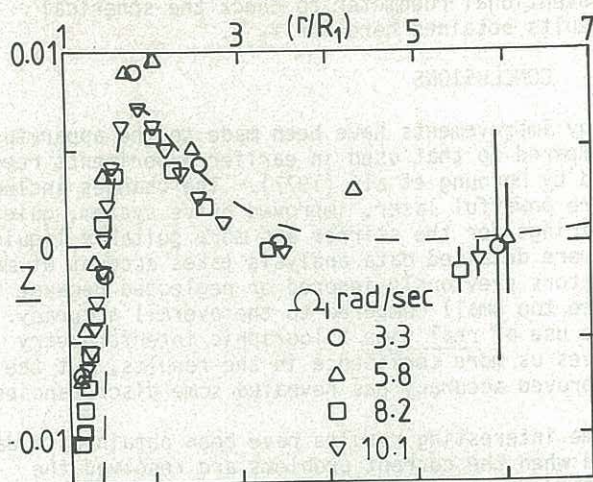


Figure 2 Fluid A spherical data, dashed line, ---, Newtonian fit.

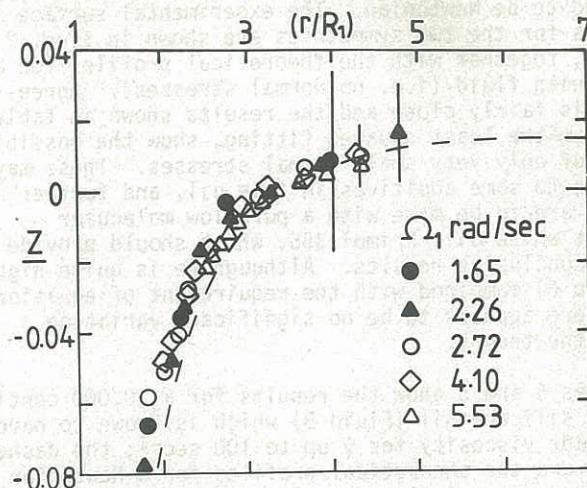


Figure 3 Fluid A cylindrical data, dashed line, ---, Newtonian fit.

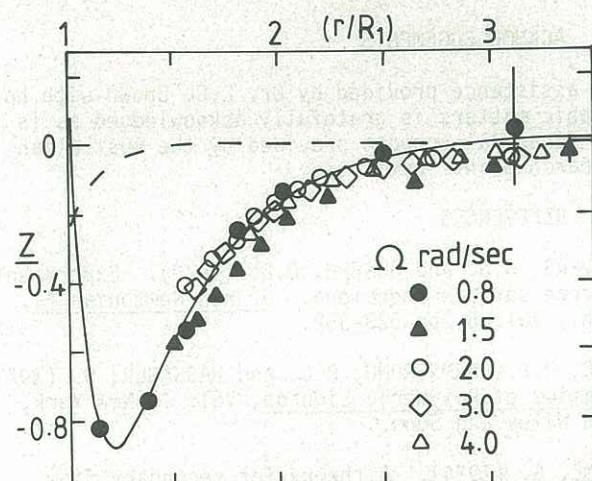


Figure 4 Fluid B spherical data, —, best fit to theory; ---, $\Psi_{1,0} = \Psi_{2,0} = 0$.

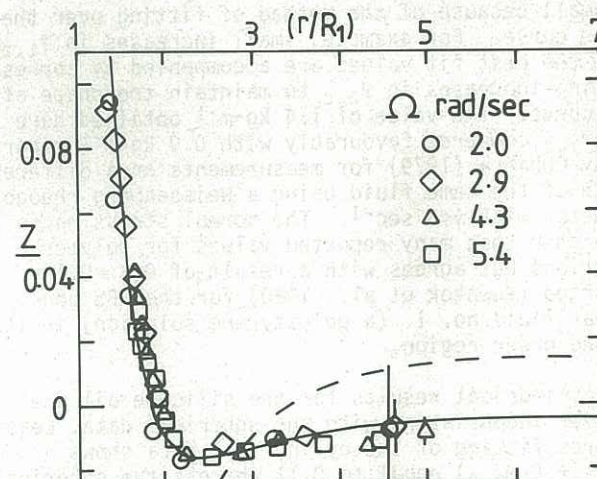


Figure 5 Fluid B cylindrical data, —, best fit to theory; ---, $\Psi_{1,0} + 4 \cdot \Psi_{2,0} = 0$.

TABLE I

FLUID PROPERTIES AT 25°C AND EXPERIMENTAL NORMAL STRESS DATA

Properties and Results	Fluid A	Fluid B
	Perfecto Medium T68 Turbine Oil Castrol Australia	Dow Corning 200 Silicone fluid 30,000 cst.
Density, ρ (kg.m^{-3})	876	975
Viscosity, η_0 (Pa.s)	0.12	32
Refractive index, n	1.4832	1.4035
Spherical data		
$\Psi_{1,0}$ (Pa.s ²)	0.024 ± 0.006	1.40 ± 0.18
$\Psi_{2,0}$ (Pa.s ²)	-0.006 ± 0.002	-0.44 ± 0.06
$-\Psi_{2,0}/\Psi_{1,0}$ (ratio)	0.25 ± 0.02	0.31 ± 0.01
Cylindrical data		
$\Psi_{1,0} + 4 \cdot \Psi_{2,0}$ (Pa.s ²)	0.023 ± 0.010	0.105 ± 0.015
Range of Reynolds no.	1.0 to 6.2	0.004 to 0.02
Range of Weissenberg no.	0.002 to 0.020	0.07 to 0.32

commercial oil (Fluid A, see table 1) which was believed to be Newtonian. The experimental surface shapes for the two symmetries are shown in figs. 2 and 3, together with the theoretical profiles for a Newtonian fluid (i.e. no normal stresses). Agreement is fairly close and the results shown in table 1, from the least squares fitting, show the possibility of only very small normal stresses. These may be due to some additives in this oil, and further tests are to be made with a pure low molecular weight white oil, Primol 355, which should provide more conclusive results. Although Re is quite high (up to 6) compared with the requirement of equation 2, there appears to be no significant variation from the theory.

Figures 4 and 5 show the results for a 30,000 centistoke silicone oil (Fluid B) which is known to have constant viscosity for $\dot{\gamma}$ up to 100 sec^{-1} ; the dashed lines are the theoretical profiles for a Newtonian liquid and are included to show the effect of elasticity on the shape of the free surface. The theoretical fit to the spherical data gives the ratio $-N_2/N_1$ as 0.31 with an error of only $\pm 3\%$. The error is small because of the method of fitting over the whole curve. For example, small increases in $\Psi_{1,0}$ from the best fit values are accompanied by corresponding increases in $\Psi_{2,0}$ to maintain the shape of the curve. The value of $1.4 \text{ kg}\cdot\text{m}^{-1}$ obtained here for $\Psi_{1,0}$ compares favourably with $0.9 \text{ kg}\cdot\text{m}^{-1}$ reported by Chhabra (1979) for measurements on a different batch of the same fluid using a Weissenberg rheogoniometer with $\dot{\gamma} < 5 \text{ sec}^{-1}$. The normal stress ratio is higher than many reported values for polymer solutions but agrees with a result of 0.3 ± 0.1 reported (Keentok et al., 1980) for the NBS non-linear fluid no. 1. (a polystyrene solution) in its second order region.

The cylindrical results for the silicone oil are however inconsistent with the spherical data. Least squares fitting of the cylindrical data shows $(\Psi_{1,0} + 4\Psi_{2,0})$ equal to 0.11 whereas the spherical data gives -0.36 for this linear combination. The reasons for this discrepancy are not fully understood. It is believed that errors in the curve fitting to the profiles obtained are not responsible for the discrepancy. The surface profiles are reproducible and similar discrepancies have been observed for other silicone oils. The effect on the results of possible lack of symmetry in the experimental apparatus have been investigated but none is very large. The values of Re and We are sufficiently small that the second order theory should apply. Note that although Z appears to be independent of Ω_1 in the figures, this is only a necessary, but not a sufficient, condition for second order behaviour. There remains the possibility that end effects in the cylindrical case are significantly disturbing the flow compared to that predicted by the theory; or that some effect on the fringes has been overlooked, although the close agreement of fluid A to the theory seems to exclude this possibility. The end effect has been partially investigated by changing the depth of the fluid in the bowl but the basic shape of the surface remains unchanged at different depths. The end effects are to be investigated further, and measurements of N_1 are to be made on a

conventional rheometer to check the spherical results obtained here for $\Psi_{1,0}$.

5 CONCLUSIONS

Many improvements have been made to the apparatus compared to that used in earlier experiments reported by Hornung et al. (1977). The changes include a more powerful laser, improved drive system, quieter bearings for the stirrer and more suitable liquids. A more detailed data analysis takes account of many factors previously ignored or neglected because they were too small compared to the overall accuracy. The use of real time holographic interferometry gives us more confidence in the results, but the improved accuracy has revealed some discrepancies.

Some interesting results have been obtained to date and when the current problems are resolved the method should provide very accurate results for the normal stress ratio N_2/N_1 at low shear rates. The accuracy of most other methods is degraded by having to combine the errors from two separate measurement techniques.

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