



# Viscometric functions of semi-dilute non-colloidal suspensions of spheres in a viscoelastic matrix



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## ABSTRACT

The viscometric functions ( $\eta$ ,  $N_1$  and  $N_2$ ) for non-colloidal suspensions of spheres in a Boger fluid matrix were measured. Volume fractions ( $\phi$ ) of 5%, 10% and 20% were investigated. The relative viscosity ( $\eta_r = \eta/\eta_0$ ) and the (positive) first normal stress difference  $N_1$  showed increases with  $\phi$  which were larger than the dilute suspension theory predictions of  $1 + 2.5\phi$ , indicating semi-dilute suspension behaviour.

The main interest centres on the second normal stress difference  $N_2$ . The matrix fluid showed a zero second normal stress difference, and the measurements showed that  $N_2$  was always negative for the suspensions. This agrees with the dilute suspension prediction found using the Landau-Lifschitz averaging procedure, but not with the ensemble averaging method, which predicts a positive  $N_2$ . Possible causes for this result are discussed.

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## 1. Introduction

Here we report the results of measuring the viscometric functions ( $\eta_r$ ,  $N_1$ ,  $N_2$ ) in semi-dilute non-colloidal suspensions of spheres in a viscoelastic Boger fluid matrix. The results are then compared with existing predictions for the dilute case from suspension theory [1–4]. The predictions are of two kinds – Koch and Subramanian [1] and Rallison [2] used an ensemble averaging procedure to find the average bulk stresses, while Greco et al. [3] and Housiadas and Tanner [4] used a direct averaging procedure based on Landau and Lifschitz's work [5]. All four predictions modelled the matrix fluid by a second-order equation [6]

$$\boldsymbol{\sigma} = -p\mathbf{I} + \eta_0\mathbf{A} + (\psi_1 + \psi_2)\mathbf{A}^2 - \frac{1}{2}\psi_1\mathbf{B} \quad (1)$$

where  $\boldsymbol{\sigma}$  is the total stress tensor,  $p$  is a pressure,  $\mathbf{I}$  is the unit tensor,  $\eta_0$  is the (constant) matrix viscosity and  $\psi_1$  and  $\psi_2$  are the (constant) first and second normal stress coefficients.  $\mathbf{A}$  is defined as twice the rate of deformation tensor:

$$\mathbf{A} = \mathbf{L} + \mathbf{L}^T \quad (2)$$

where  $L_{ij} = \partial v_i / \partial x_j$  are the components of the velocity gradient tensor  $\mathbf{L}$ ;  $v_i$  is the velocity component in the  $i$ -direction.

The second Rivlin-Ericksen tensor  $\mathbf{B}$  is defined as [6]

$$\mathbf{B} = \frac{D\mathbf{A}}{Dt} + \mathbf{L}^T\mathbf{A} + \mathbf{A}\mathbf{L} \quad (3)$$

where the particle – following derivative  $D/Dt = \partial/\partial t + \mathbf{v} \cdot \nabla$ .

The matrix fluid is assumed to be incompressible. In a simple shear flow (velocity field  $\mathbf{v} = \dot{\gamma}y\mathbf{i}$ ), the normal stress differences  $N_1(0)$  and  $N_2(0)$  in the matrix fluid are given by

$$N_1(0) \equiv \sigma_{xx} - \sigma_{yy} = \psi_1\dot{\gamma}^2 \quad \text{and} \quad N_2(0) \equiv \sigma_{yy} - \sigma_{zz} = \psi_2\dot{\gamma}^2. \quad (4)$$

The predictions [1–4] all relate to the changes in the viscometric functions as rigid spherical particles are added to the viscoelastic matrix. To the first order in the volume fraction of spheres ( $\phi$ ) the results are as follows:

For the viscosity  $\eta(\phi)$  and the first normal stress difference  $N_1(\phi)$ , they are given by [1–4]

$$\eta(\phi) = \eta_0(1 + 2.5\phi) \quad (5)$$

and

$$N_1(\phi) = N_1(0)(1 + 2.5\phi) \quad (6)$$

respectively.

For the second normal stress difference  $N_2(\phi)$ , the predictions are different: one finds [3,4]

$$N_2(\phi) = N_2(0) \left[ 1 + \left( \frac{10}{7} - \frac{45}{56} \frac{N_1(0)}{N_2(0)} \right) \phi \right] \quad (7)$$

and from [1,2,7]

$$N_2(\phi) = N_2(0) \left[ 1 + \left( \frac{75}{28} + \frac{5}{56} \frac{N_1(0)}{N_2(0)} \right) \phi \right] \quad (8)$$

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There are few, if any, suitable measurements to compare with these dilute suspension results. The work by Mall-Gleissle et al. [8] used semi-dilute suspensions of spheres (down to 5% volume fraction) in silicone matrices and it appears that the relation formed by combining (5) and (6), which is [9]

$$\frac{N_1(\phi)}{N_1(0)} = \eta_r \tag{9}$$

is obeyed by these results even though the Weissenberg number ( $Wi \equiv \lambda \dot{\gamma}$ ) is of order 1 or greater for the flows considered. It is however debatable if the second-order model should be used unless  $Wi < 1.0$ . The increase of the viscosity with concentration is more rapid than Eq. (5) suggests. Unfortunately the data on the second normal stress difference are too scattered to distinguish between (7) and (8), although  $N_2$  was always negative. Therefore new experiments, following the suggestion of Rallison [2], have been made and are reported here.

**2. Experiments**

It is necessary to determine  $N_2$  accurately, and the best choice appears to be the semi-circular open channel [10,11] where  $N_2$  can be measured to around 0.1 Pa accuracy. In this test, fluid flows down an inclined semi-circular trough under gravity and the free surface deflection is viewed optically. Fig. 1 shows the cross-section of the trough and the elevation  $h(u)$  that is measured via the reflection of a millimetre scale in the free surface. Once  $h$  has been determined, then the second normal stress difference can be found [6,10,11] from the formula

$$-N_2 = \frac{1}{u} \int_0^u x dQ \tag{10}$$

where

$$Q(x) = \rho gh(x) \cos \beta + \frac{\gamma_s}{\rho^*} \tag{11}$$

and  $\rho$  is the fluid density,  $g \cos \beta$  is the component of the gravitational acceleration ( $g$ ) normal to the tube axis,  $\beta$  is the angle of the tube axis to the horizontal (here  $\beta$  was  $30^\circ$  for all tests),  $\gamma_s$  is the surface tension coefficient (here taken as 0.07 Pa m) and  $\rho^*$  is the radius of curvature of the free surface (Fig. 1). The shear stress ( $\tau$ ) in the tube increases linearly from the centreline to the tube wall, and has the magnitude

$$\tau \sim \frac{1}{2} \rho g R \sin \beta. \tag{12}$$

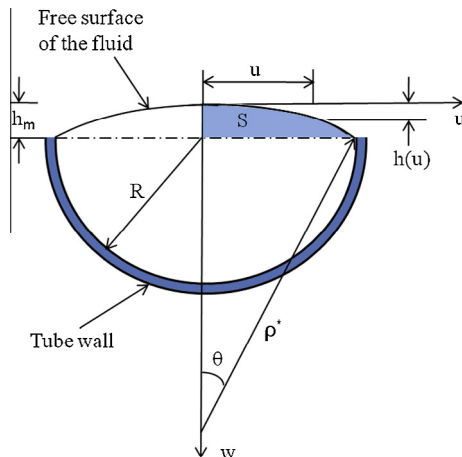


Fig. 1. Showing the definitions used to define the surface profile in the open trough cross-section. If  $h_m$  is positive as shown, then  $N_2$  is negative.

There is a small correction of  $O(h_m)$  to Eq. (12) due to the movement of the free surface (shaded area  $S$  in Fig. 1) [11] which has been taken into account in the data reduction. Note that if  $h > 0$ , then  $N_2 < 0$ ; when  $h = 0$ , then  $\rho^*$  is very large, and  $N_2 = 0$ .

We used 40  $\mu\text{m}$  diameter PMMA spheres from MICROBEADS™ as particles with a density of 1.2 g/ml, and the matrix fluid was a mixture of corn syrup (by weight 79.42%), glycerin (19.8%), water (0.75%) and a small amount (0.03%) of PAA gave it the needed non-Newtonian properties. The mixture is a Boger fluid [12] with a nearly-constant viscosity and a density of 1.352 g/ml. The Péclet number ( $Pe$ ) is large ( $>10^8$ ) and so the suspensions are non-colloidal.

From the trough experiment (Fig. 2), no surface deflection with the matrix fluid was detected, indicating that  $N_2(0) \approx 0$  for the matrix. Hence the matrix behaves as a Boger fluid with  $N_2(0) = 0$ . This is important because if  $N_2(0) = 0$ , then the two results (7) and (8) reduce to the Koch-Rallison [1,2,7] result

$$N_2(\phi) = + \frac{5}{56} N_1(0) \phi \tag{13}$$

and the Greco-Housiadas result [3,4]

$$N_2(\phi) = - \frac{45}{56} N_1(0) \phi. \tag{14}$$

Hence there is a difference in sign between these results – a positive  $N_2$  is predicted for one case and a negative  $N_2$  for the other. These correspond respectively to a depression of the surface (positive  $N_2$ ) and an arching up of the surface (negative  $N_2$ ) [6,9,10], if  $N_1(0)$  is positive. A parallel-plate rheometer (Paar Physica MCR301) was used to find  $N_1 - N_2$  and the relative viscosity  $\eta(\phi)/\eta_0$ . The standard methodology for the deduction of  $N_1 - N_2$  and the viscosity from the measured normal thrust and torque data is set out in references [6] and [11]. With the parallel-plate system we have, since  $N_2 = 0$  here

$$N_1 = f \left[ 2 + \frac{d \log f}{d \log \dot{\gamma}_0} \right] \tag{15}$$

where  $f = F/\pi R^2$ . Here  $F$  is the normal thrust on the plates, radius  $R$ , and  $\dot{\gamma}_0$  is the shear rate at the rim. The viscosity follows from

$$\eta(\dot{\gamma}_0) = \frac{m}{\dot{\gamma}_0} \left[ 3 + \frac{\log m}{\log \dot{\gamma}_0} \right] \tag{16}$$

where  $m = M/2\pi R^3$ ; here  $M$  is the torque on the plates. The results for the matrix are shown in Fig. 3. The matrix viscosity varied slightly over the range of shear stresses encountered in the trough (up to about 40 Pa); there is about a 6% reduction at  $\dot{\gamma} = 35 \text{ s}^{-1}$  from the zero-shear rate viscosity (2.22 Pa s), and the average viscosity is

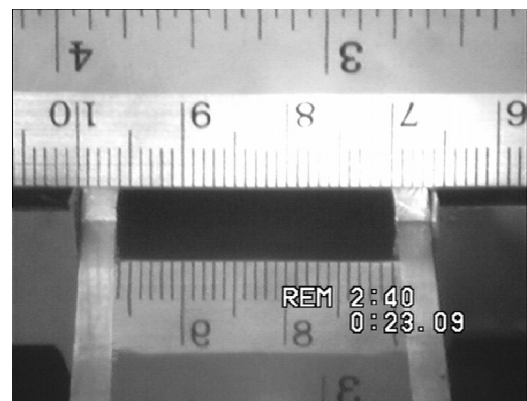


Fig. 2. A snapshot shows that the matrix fluid surface is flat in the tube channel flow, implying that  $N_2 \sim 0$  for this material.

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