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## STATIC SHEAR VISCOSITY OF A BIMODAL SUSPENSION

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The static shear viscosity  $\eta$  of a bimodal suspension is studied. The value and volume fraction dependence of  $\eta$  is determined within the cell approach. It is shown that the characteristic peculiarities in the behaviour of  $\eta$  are connected with inhomogeneous distribution of small disperse particles in a system. The comparison of the obtained results with the computer simulation data is performed.

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### 1. Introduction

The rheological properties of bimodal suspensions have numerous technical applications [1]. This circumstance stimulated experimental [2] theoretical [5] studies and computer simulations [3, 4] in the last years. The main attention of the investigators was focused on the dependence of the shear viscosity on the volume fractions  $\Phi_1$  and  $\Phi_2$  occupied by components and the ratio  $\lambda = \frac{R_1}{R_2}$  of their radii. In the experiments [2, 6] and computer simulations [3] the following characteristic details of the shear viscosity  $\eta(\zeta, \lambda)$  considered as a function of  $\zeta = \frac{\Phi_1}{\Phi}$  at constant  $\Phi = \Phi_1 + \Phi_2$  and  $\lambda$  were discovered: 1)  $\eta(0, \lambda) = \eta(1, \lambda)$ ,  $\eta(\zeta, \lambda) < \eta(0, \lambda)$ , i.e. the shear viscosity of a bimodal suspension is always less than  $\eta$  for a one-component suspension with the same volume fraction  $\Phi$ ; 2) the decrease of  $\eta(\zeta, \lambda)$  at  $\lambda = \text{const}$  near  $\lambda = 0$  and  $\lambda = 1$  is asymmetric:

$$|\eta'_\zeta(0, \lambda)| \neq \eta'_\zeta(1, \lambda);$$

3) the minimum value  $\eta_m(\lambda) = \min \eta(\zeta, \lambda)$  of the shear viscosity increases with  $\lambda$ .

Such a behaviour of  $\eta(\zeta, \lambda)$  was an object of the theoretical analysis in works [2, 5]. In [2], the expression

$$\eta(\zeta, \lambda) = \eta_0 \left( 1 - \frac{\Phi}{\Phi_m(\zeta, \lambda)} \right)^{-0.15}$$

was proposed. It has the heuristic character and cannot be justifying by the hydrodynamic methods in their applicability region.

In the present work, we study the shear viscosity of a bimodal suspension in terms of the cell approach developed in [7, 8] and appropriate physical arguments. In such a way, we are able to calculate the shear viscosity for the following cases: 1) not dense suspension ( $\Phi < 0.15$ ); 2) dense suspension ( $\Phi < 0.5$ ) with  $\lambda \gg 1$  and 3) suspension with close components ( $\lambda \sim 1$ ). It will be shown that the results obtained are in a quite satisfactory agreement with the experimental data.

### 2. Shear Viscosity of Monodisperse Suspension

To describe the average shear viscosity  $\eta$  of a one-component suspension of spherical particles, we will use the cell approach developed in [8]. The distinctive features of the last are: 1) the rotational motion of a particle in the spherical cell is considered; 2) the dependence of the cell radius on the volume fraction  $\Phi$  occupied by particles is taken into account. Due to the first point, the symmetry of hydrodynamic flows is consistent with the shape of a cell. The second assumption allows us to choose the values of the model parameters that reproduce, at small  $\Phi$ , the first terms of

the  $\eta$ -expansion in the series with respect to  $\Phi$  obtained with help of the hydrodynamic perturbation theory [9]. In such a way, the following expression for the average shear viscosity has been found [8]:

$$\eta = \eta_0 F(\psi). \quad (1)$$

Here,  $\eta_0$  is the shear viscosity of a liquid,  $\psi = \left(\frac{R_p}{R_C}\right)^3$ ,  $R_p$  and  $R_C$  are the radii of a particle and the cell, correspondingly,

$$F(\psi) = \frac{\psi(1-\psi)}{1 + \psi(1-\psi) - \sqrt{1 + 2\psi^2(1-\psi)}}. \quad (2)$$

The dependence of  $R_C$  on  $\Phi$  is given by the formula

$$R_C = a(\Phi)R_G, \quad a(\Phi) = \alpha_0 + \alpha_1\Phi + \alpha_2\Phi^2 \dots, \quad (3)$$

where  $R_G$  is the average interparticle distance,

$$\alpha_0 = \left(\frac{6}{2.5\pi}\right)^{\frac{1}{3}} \approx 0.93,$$

$$\alpha_1 \approx \frac{\pi\alpha_0^4}{18} \left[ \frac{6}{\pi\alpha_0^3} - 5.2 \right] \approx 0.127, \quad \alpha_2 \approx 0.03. \quad (4)$$

It is evident that

$$\Phi = \frac{\pi}{6} \left( \frac{R_p}{R_G} \right)^3, \quad (5)$$

and the relation between  $\psi$  and  $\Phi$  is  $\Psi = \frac{6}{\pi a^3(\Phi)} \Phi$ .

In accordance with the above, at small  $\Phi$ ,

$$F(\psi) = 1 + 2.5\Phi + 5.2\Phi^2 + \dots, \quad (6)$$

which coincides with the expression given in [9].

The hydrodynamic description of the rotational motion of a particle inside the cell is correct up to  $\Phi_x \approx 0.49$ , corresponding to the occasional dense packing of spherical particles [10]. The comparison with experimental data [11] shows that, in the interval  $0 < \Phi < 0.49$ , there is a very good agreement between them and the values calculated according to (1) with parameters (3)–(5).

It is necessary to emphasize that the shear viscosity of a one-component suspension is only determined by the volume fraction of suspended particles and does not depend on their radius. This conclusion correlates successfully with experimental data [11]. The dependence of  $\eta$  on  $R_p$  is expected for  $\Phi > \Phi_x$  only.

### 3. Bimodal Suspension with Strongly Different Particles ( $R_2 \ll R_1$ ) in the Case where $\Phi_1 \ll \Phi_2 \leq \Phi$

In this Section, we consider the shear viscosity of a bimodal suspension, whose fractions differ from each other by the radii ( $R_2 \ll R_1$ ) of their particles and the volume fractions  $\Phi_1$  and  $\Phi_2$  satisfying the inequality  $\Phi_1 \ll \Phi_2$ . Obviously, the small disperse fraction can be considered as a suspending fluid for the second fraction. Therefore, we can write

$$\eta = \eta_2 F(\psi_1), \quad (7)$$

where the meaning of  $\psi_1$  is evident. In its turn, the shear viscosity of the small disperse fraction of the suspension can be approximated by formula (1):

$$\eta_2 = \eta_0 F(\psi_2). \quad (8)$$

Hence,

$$\eta = \eta_0 F(\psi_1) F(\psi_2). \quad (9)$$

Let  $\varphi_2$  be the volume fraction of the small disperse phase. It is connected with  $\Phi_2$  by the relation

$$\varphi_2 = \frac{\Phi_2}{1 - \Phi_1}. \quad (10)$$

Since

$$\psi_2 = \frac{6}{\pi a^3(\varphi_2)} \varphi_2 \quad (11)$$

and

$$\Phi = \Phi_1 + \Phi_2, \quad (12)$$

relation (9) yields

$$\begin{aligned} \eta(\Phi_1, \Phi) &= \eta_0 \chi(\Phi_1) \chi\left(\frac{\Phi - \Phi_1}{1 - \Phi_1}\right) \equiv \\ &\equiv \eta \chi(\zeta \Phi) \chi\left(\frac{1 - \zeta}{1 - \zeta \Phi} \Phi\right), \end{aligned} \quad (13)$$

where  $\chi(t) = F\left(\frac{6}{\pi a^3(t)} t\right)$ . The applicability region of (13) is determined by the inequality

$$\Phi_1 \ll \Phi_2 \leq \Phi < 0.5. \quad (14)$$

If  $\Phi \leq 0.15$ , then one can use, in accordance with (6), the expression

$$\begin{aligned} \eta(\Phi_1, \Phi) &= \\ &= \eta_0 [1 + 2.5\Phi + 5.2\Phi^2 - (1.65\Phi - 23.5\Phi^2)\zeta\Phi + \dots]. \end{aligned} \quad (15)$$

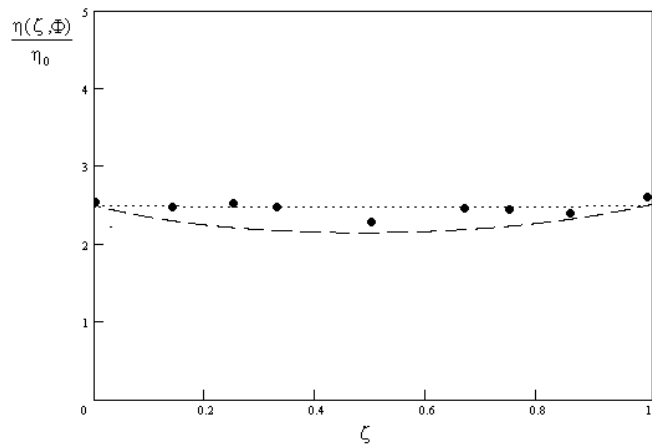


Fig. 1. Dependence of the shear viscosity of a bimodal suspension on  $\zeta$  at volume fraction  $\Phi = 0.3$  and  $\lambda = 5$ . Dotted and dashed lines correspond to Eqs. (13) and (22), respectively, circles are experimental data [2]

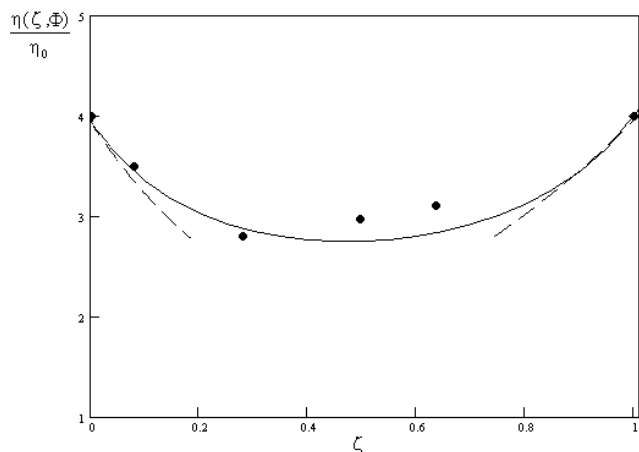


Fig. 2. Dependence of the shear viscosity of a bimodal suspension on  $\zeta$  at volume fraction  $\Phi = 0.4$ . Solid line corresponds to (21), dash lines show the asymptotic behavior of  $\eta$  at  $\zeta \rightarrow 0$  (left line) and  $\zeta \rightarrow 1$  in accordance with (19) and (18), circles are computer simulation data [3] for the same  $\Phi$  and  $\lambda = 4$

As we see, the exchange of some part of small disperse particles by large disperse ones of the same volume fraction leads to a decrease in the shear viscosity.

Formula (13) is also qualitatively applicable in the limiting case  $\Phi_1 \rightarrow \Phi$  and in the intermediate region  $\Phi_1 \sim \Phi_2$ . It is not difficult to verify that (13) leads to the following consequences:

$$\eta(0, \Phi) = \eta(\Phi, \Phi) \quad (16)$$

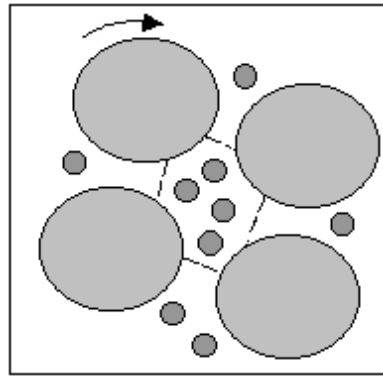


Fig. 3. Character of an inhomogeneous distribution of small particles in a system with rotating large particles

and

$$\left| \frac{\eta'_\zeta(0, \Phi)}{\eta'_\zeta(\Phi, \Phi)} \right| = 1 - \Phi. \quad (17)$$

This means that changing the shear viscosity for the left side of the interval  $0 \leq \Phi_1 \leq \Phi$  should be more sloping in comparison with that for the right side of the interval. The curve corresponding to (13) is in a satisfactory agreement with the experimental data [2] for  $\Phi = 0.3$  and  $\lambda = 5$  (see Fig. 1). However, as follows from Fig. 2, the agreement worsens for  $\Phi = 0.4$  and  $\lambda = 4$ .

#### 4. Influence of the Inhomogeneous Distribution of Small Particles and Inertial Effects

The results obtained in Section 3 correspond to the assumption about the homogeneous spatial distribution of both disperse fractions. This condition seems to be quite correct only at  $\Phi_1 \ll \Phi_2 < \Phi$ . At  $\Phi_1 \sim \Phi_2$  for dense enough suspensions,  $0.2 \leq \Phi \leq 0.5$ , the homogeneous distribution of the small dispersed fraction is violated. This effect is particularly pronounced in the limiting case  $\Phi_2 \ll \Phi_1 \ll \Phi$ . To diminish the production of entropy caused by the rotation of large particles, small particles should be accumulated in the regions (voids) maximally removed from the centers of large particles (see Fig. 3). In these regions, the gradients of the velocity field take minimal values. This conclusion is not related to the specificity of the rotational motion. In this situation, the shear viscosity of a bimodal suspension is mainly determined by the large disperse fraction:

$$\eta(\Phi_1, \Phi) \approx \eta_0 \chi(\Phi_1) = \eta_0 \chi(\zeta \Phi), \quad \zeta \rightarrow 1. \quad (18)$$

The stronger the inequality  $\Phi_2 \ll \Phi_1$ , the higher the accuracy of this relation is.

Some revision of formula (13) is also necessary in the opposite limiting case  $\Phi_1 \ll \Phi_2 \ll \Phi$ . Here, we should take into account that the mobility of large particles is considerably less in comparison with that of small particles. Due to this, the shear viscosity of the system is mainly formed by small particles:

$$\eta(\Phi_1, \Phi) \approx \eta_0 \chi(\varphi_2). \quad (19)$$

Here, in accordance with (10),

$$\varphi_2 = [1 - (1 - \Phi)\zeta]\Phi, \quad \zeta \ll 1. \quad (20)$$

The difference between the values of  $\eta(\zeta, \Phi)$  given by formulas (13) and (19) is proportional to  $\zeta$ :

$$\Delta\eta(\Phi_2, \Phi) = 2.5\zeta\eta_0\Phi\chi([1 - (1 - \Phi)\zeta]\Phi).$$

With increase in  $\zeta$ , this effect becomes less pronounced.

Thus, we expect at  $\Phi_1 \sim \Phi_2$  that the shear viscosity will be less than its value given by (13). The rotation of large particles leads to the redistribution of small ones so that the effective value of the volume fraction  $\varphi_2^{(\text{eff})}$  diminishes in comparison with  $\varphi_2 = \frac{1-\zeta}{1-\zeta\Phi}\Phi$ . Since the accumulation effect in (18) increases with  $\Phi_1 = \zeta\Phi$ , one can assume that the effective volume fraction  $\varphi_2^{(\text{eff})}$  decreases:

$$\varphi_2^{(\text{eff})} = \varphi_2(1 - \zeta\Phi)^p. \quad (21)$$

The value of  $p$  seems to be natural to identify with that determining the production  $\delta S$  of entropy as a function of the gradient velocity  $g_{ik}$ :  $\delta S \sim \int_V dV g_{ik}^2$  ( $V$  is the volume of a cell). This yields that  $p = 2$ .

As a result, the shear viscosity of a bimodal suspension is assumed to be equal to

$$\eta = \eta_0(\zeta\Phi)\chi((1 - \zeta)(1 - \zeta\Phi)\Phi). \quad (22)$$

The limit values of  $\eta(\Phi_2, \Phi)$  at  $\Phi_2 = 0$  and  $\Phi_2 = \Phi$  are the same:

$$\eta(0, \Phi) = \eta(\Phi, \Phi) = \eta_0\chi(\Phi). \quad (23)$$

However, their behavior is asymmetric at  $\Phi_2 \rightarrow 0$  and  $\Phi_2 \rightarrow \Phi$ :

$$\begin{aligned} \eta(\Phi_2, \Phi) &= \\ &= \begin{cases} \eta(0, \Phi) - \chi'(\Phi)\Phi(1 - \zeta), & \zeta \rightarrow 0, \\ \eta(0, \Phi) - \chi'(\Phi)(1 - \Phi)(\Phi - \Phi(1 - \zeta)), & \zeta \rightarrow 1. \end{cases} \end{aligned} \quad (24)$$

Thus, it follows from our analysis that the decay rates of the shear viscosity near the left and right points of the interval  $(0, \Phi)$  are related as

$$\sigma = \left| \frac{\eta'_\zeta(1, \Phi)}{\eta'_\zeta(0, \Phi)} \right| = 1 - \Phi. \quad (25)$$

This result correlates with experimental data [2] quite satisfactorily (see Fig. 2). There, the comparison of the experimental and theoretical values of  $\eta(\zeta, \Phi)$  calculated according to (22) is also presented. It testifies to the favor of our assumption about the inhomogeneous distribution of small particles.

## 5. Bimodal Suspension of Components with $R_1 \approx R_2$

Let us consider a bimodal suspension, whose components differ slightly from each other only by their radii:  $R_1 \approx R_2$ . To determine the shear viscosity of such a system, we generalize the cell approach developed in [8]. More exactly, we complete it by the arguments characteristic of the mean field approximation.

Firstly, let  $\bar{R}$  and  $R_C$  be the average radii of particles and the spherical cell:

$$\bar{R}^3 = P_1 R_1^3 + P_2 R_2^3, \quad (26)$$

$$R_C^3 = P_1 R_{C1}^3 + P_2 R_{C2}^3, \quad (27)$$

where

$$P_1 = \frac{n_1}{n_1 + n_2} = \frac{\Phi_1}{\Phi_1 + \lambda^3 \Phi_2}$$

is the probability to take a particle of the first type,  $P_2 = 1 - P_1$ ,  $\lambda = \frac{R_1}{R_2}$ ,  $n_i$ ,  $i = 1, 2$ , are the numerical densities of particles.

The relation between the radii of particles and the corresponding spherical cell is set by the formulas

$$R_{C1}^3 = \frac{\pi a^3(\Phi)}{6\Phi} R_1^3, \quad R_{C2}^3 = \frac{\pi a^3(\Phi)}{6\Phi} R_2^3 \quad (28)$$

similarly to that in [8], where  $a(\Phi)$  is given by (3).

To determine the shear viscosity of a bimodal suspension, we use the equation of energetic balance (see [8])

$$W(\bar{R}, \eta) = P_1 W(R_1, \eta_0, \eta) + P_2 W(R_2, \eta_0, \eta), \quad (29)$$

where  $W(R_1, \eta_0, \eta)$  is the energy dissipation rate in a cell occupied by the first particle,  $\eta_0$  and  $\eta$  are the shear viscosities of the suspending fluid and the bimodal suspension,  $W(R_2, \eta_0, \eta)$  takes the analogous meaning,  $W(\bar{R}, \bar{\eta})$  is the energy dissipation rate for an

isolated particle of the average radius immersed in a homogeneous liquid with viscosity  $\bar{\eta}$ .

According to [8], we get

$$W_1(R_1, \eta_0, \eta) = \frac{8\pi}{3} \frac{(1 - \psi_1)(2 + \psi_1 z^2)}{(1 - \psi_1 z)^2} \Omega^2 R_1^3 \eta_0, \quad (30)$$

$$W_2(\bar{R}, \eta) = \frac{16\pi}{3} \left( \frac{1 - z}{1 - \psi z} \right)^2 \Omega^2 \bar{R}^3 \eta, \quad (31)$$

where  $z = 1 - \frac{\eta_0}{\eta}$ ,  $\psi_1 = \frac{R_1^3}{R_{C1}^3}$ ,  $\psi_2 = \frac{R_2^3}{R_{C2}^3}$ ,  $\psi = \frac{6\Phi}{\pi a^3(\Phi)}$ ,  $i = 1, 2$ .

After respective substitutions, Eq. (29) transforms to

$$\begin{aligned} & 2 \frac{1 - z}{(1 - \psi z)^2} \Phi = \\ & = \frac{(1 - \psi_1)(2 + \psi_1 z^2)}{(1 - \psi_1 z)^2} \Phi_1 + \frac{(1 - \psi_2)(2 + \psi_2 z^2)}{(1 - \psi_2 z)^2} \Phi_2 \end{aligned} \quad (32)$$

and is, in fact, the equation for  $z$ . In the approximation linear in the small parameter  $\delta = \lambda^3 - 1 \ll (<)1$ , we find

$$z = z_0 - \delta(3.45 + 2.34\psi), \quad (33)$$

where

$$z_0 = \frac{-1 + \sqrt{1 + z\psi^2(1 - \psi)}}{\psi(1 - \psi)}.$$

Thus, for the average shear viscosity of a bimodal suspension, we get

$$\eta(\psi, \delta) = \eta_0 [\chi(\psi) - \delta \chi^2(\psi)(3.45 + 2.34\psi)] + O(\delta^2). \quad (34)$$

It is very essential that  $\eta$  is a function of  $\Phi$  and  $\delta$  only, which agrees fully with the computer simulation results [3]. The correction term in (34) proportional to  $\delta$  is negative, which also corresponds to the tendency observed in the laboratory and computer experiments [3].

## 6. Conclusion

In the present work, we have studied the behaviour of the shear viscosity  $\eta$  of a bimodal suspension. To describe the dependence of  $\eta$  on the dimensionless parameter  $\zeta = \frac{\Phi_1}{\Phi}$  and  $\lambda = \frac{R_1}{R_2}$ , the modified version of the cell approach [8] is used. It is shown that, at  $\Phi = 0.3$  and  $\lambda > (\gg)1$ , the dependence of  $\eta$  on  $\zeta$  is quite weak. In this connection, the change of  $\eta$  at  $\zeta \rightarrow 1$  is more essential in comparison with that for  $\zeta \rightarrow 0$ , that is in agreement with the results of Section 4. Unfortunately, it is difficult to obtain more detailed numerical estimates because of the small number of the experimental points.

It is taken into account that the spatial distribution of small particles is inhomogeneous for dense enough suspensions. To diminish the production of entropy in the system, the particles with smaller radii should be accumulated in the regions with minimal velocity gradients. Due to this fact, the dependence of  $\eta(\zeta, \Phi)$  near  $\zeta \rightarrow 0$  and  $\zeta \rightarrow 1$  is opposite to that corresponding to the case  $\Phi = 0.4$ .

The experimental results obtained in [2] are in a satisfactory agreement with this conclusion. In particular, at  $\Phi = 0.4$  and  $\lambda = 4$  the ratio  $|\eta'_\zeta(0, \Phi)|/|\eta'_\zeta(1, \Phi)|$  of the derivatives of the shear viscosity at  $\zeta \rightarrow 0$  and  $\zeta \rightarrow 1$  is close to  $(1 - \Phi)$ , as follows from (24). This fact supports our arguments given in Section 4 about the inhomogeneous distribution of small particles around large ones.

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## СТАТИЧНА ЗСУВНА В'ЯЗКІСТЬ БІМОДАЛЬНОЇ СУСПЕНЗІЇ

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### Резюме

Вивчено статичну зсувну в'язкість  $\eta$  бімодальної суспензії. Отримано значення  $\eta$  та її залежність від питомого об'єму за допомогою коміркового підходу. Показано, що характерні особливості у поведінці  $\eta$  пов'язані з неоднорідним розподілом малих сфер у системі. Наведено порівняння отриманих результатів з експериментальними даними.